PB 184751

CHEMICAL RESEARCH AND DEVELOPMENT CENTER

FMC CORPORATION

PRINCETON, NEW JERSEY

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PCR-684

APPLICABILITY OF INORGANIC SOLIDS OTHER THAN OXIDES TO THE DEVELOPMENT OF NEW PROCESSES FOR REMOVING SO_2 FROM FLUE GASES

Final Phase I Report
Contract PH-22-68-57

for

New Process Development Section
Division of Process Control Engineering Program
National Air Pollution Control Administration
Public Health Service
U. S. Department of Health, Education and Welfare

FMC Corporation
Chemical Research and Development Center
Central Research Department

June, 1969

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INTRODUCTION

The magnitude of problems associated with pollution control has commanded considerable attention during the last few years. The Federal Government currently is engaged in a program to develop acceptable methods to control emissions from stationary air pollution sources. Until now the discharge of pollutants into the atmosphere has been a common practice. Now these discharges have reached such proportions that emission control has become a matter of considerable concern. Major gaseous contributors to atmospheric pollution are hydrocarbons, carbon monoxide, nitrogen oxides, and sulfur oxides from the combustion of fossil fuels and hydrocarbons.

The emission of sulfur oxides into the atmosphere is increasing rapidly and in many areas has already reached serious proportions. In large concentrations, sulfur dioxide is toxic. In lower concentrations it acts as an irritant to the respiratory tract, and prolonged exposure to as little as ten parts per million can be harmful to the elderly and those suffering from lung or circulatory diseases. Vegetation is especially sensitive to sulfur dioxide, and certain plants have been observed to show the effects of sulfur dioxide poisoning in atmospheres containing less than one part per million of sulfur dioxide. Sulfur dioxide and its oxidation product, sulfuric acid, are the principal causes of acidity in rain water and fog; and the acid is one of the major pollutants in mine drainage waters. The harmful effects of sulfur dioxide include corrosion of ferrous and non-ferrous metals, and deterioration of materials and fabrics, such as wool, cotton and leather.

The U.S. Public Health Service has estimated that 21 million tons of sulfur dioxide are released annually to the atmosphere in the United States from the combustion of coal and fuel oil (400). About 85% of this results from the combustion of coal containing from 0.5 to more than 5% of sulfur. The remaining 15% comes from the combustion of fuel oils containing less than 5% of sulfur. The fraction of sulfur in the fuel that is converted to sulfur dioxide depends on the type of fuel burnt and the method of firing used. Thus, in stoker firing of coal, 60% to 75% of the sulfur is converted to sulfur dioxide, whereas over 95% of the sulfur in coal is oxidized to sulfur dioxide in pulverized fuel firing. With oil firing, virtually all the sulfur is released as sulfur dioxide.

The largest source of sulfur oxide emission is stationery power generation plants (388, 389) which release about 46% of all sulfur oxides emitted to the atmosphere. An additional 32% comes from industrial, commercial, and miscellaneous combustion operations; 12% from the smelting of ores; and 10% from combined industrial sources, which include sulfuric acid manufacturing, pulp and paper mills, chemical plants, refuse incineration, oil refining, and coke processing. Combustion of fuel oil and coal in power plant installations generally produces flue gases containing from 0.05%

to 0.35% of sulfur dioxide. Some smelting operations produce flue gases having as much as 6% to 8% sulfur oxides. The latter streams can be partially cleaned by scrubbing solutions, such as aqueous ammonia or aluminum sulphate, and various amine solutions. The scrubbed gases often contain more SO_2 than power plant flue gases. The pollution caused by smelters, contact acid plants, and refineries is small compared to that caused by power plants and is, in most instances, concentrated in only a few localities. On the other hand, pollution caused by power plant installations is widespread. An effective clean-up of this gas with its relatively low SO_2 content has not been achieved economically to date.

It has been estimated that the electric power generating capacity of this country in 1980 will be more than double that existing in 1965. A substantial portion of this projected growth is expected to be provided by nuclear fuel power plants which emit no sulfur oxides. The majority of the remainder of the growth will be provided by fossil fuel plants. For example, projections show that coal use for power generation is expected to increase from 272 million tons in 1967 to 440 million tons in 1980 - with allowance for an optimistic growth rate of nuclear power. If such growth in electric generating capacity occurs, effective measures will have to be taken to reduce the quantity of SO_2 emitted to the atmosphere or the pollution problem will be seriously aggravated in the near future.

There are two major ways to limit pollution by sulfur dioxide from fossil fuels. The sulfur can either be removed from the fuel prior to combustion, or the sulfur dioxide can be removed from the products of combustion. The use of low sulfur fuels is not feasible. The United States does not have adequate supplies of low sulfur coal for use in power generation plants, and most of the available low sulfur coal is located in areas far removed from the major power-generating areas.

Oil desulfurization, usually via hydrogenation, is expensive and may, in some cases, make the cost of the desulfurized fuel prohibitive for power plant generation.

To cope with the problems relating to pollution from combustion sources, the NAPCA is sponsoring a series of programs aimed at studying processes for controlling the emission of sulfur oxides from power generation. Their programs are designed to promote intensive searches for new sulfur control processes having long-range potential effectiveness at reasonable costs to a greater extent than processes currently available. Controls of sulfur emission are desired for both power plants and other sources. One phase of this overall program involves the development of new processes aimed primarily at providing second generation processes for removing oxides of sulfur from flue gases resulting from the combustion of fossil fuels containing sulfur. The need for such new processes is indicated by the economics, efficiency, and by-product limitations of such first generation processes now being studied as limestone sorption and adsorption, and the alkalized alumina processes. As a part of the NAPCA new process development program, the FMC Corporation made a literature study to assess the possible utilization of non-oxide inorganic solids for sulfur dioxide removal processes. The results of this study conducted under Public Health Contract No. 22-68-57, are reported herein.

SCOPE OF WORK

Work conducted under this contract included a literature survey on the use of inorganic solids other than metal oxides for SO_2 removal, a review of existing processes which fell within the definition of the study, and thermodynamic calculations designed to study the feasibility of reactions suggested for future study.

The literature survey included the following:

1. Assembling data on the kinetics and equilibrium of sorption of SO_2 and SO_3 on various sorbants.

2. Surveying available data on the catalytic oxidation of SO_2 to SO_3 on inorganic solids other than dry metal oxides; under conditions representative of industrial stack gases and at temperatures where the sulfuric acid formed remained on or within the catalyst as a condensed phase.

3. Reviewing the chemistry and kinetics of reactions that might possibly be used to regenerate spent sorbents. Such reactions included thermal dissociation, oxidation, and reduction.

4. Surveying the chemistry and kinetics of the thermal and/or catalytic conversion of regenerated gas streams to more useful sulfur-containing compounds, and particularly to elemental sulfur.

5. Surveying the literature for materials of construction to be specified for use in the presence of gas streams prevailing in stack gases or likely to be produced in the regeneration of SO_2 sorbants.

6. Studying analytical and laboratory methods used in SO_2 removal investigations and processes. Such studies were made to obtain a clear evaluation of the data and to obtain possible information helpful in proceeding in subsequent phases of this program.

All SO_2 removal processes described in the literature which fell within the definition of this study and which had been studied in pilot plant, semi-commercial, or commercial installations, were reviewed critically. This was done to acquire a better background into the problem and difficulties that would be encountered in developing such processes, and to obtain a full understanding of the data needed to develop satisfactory processes.

Pertinent thermodynamic data located in the literature and those received from Tracor, Inc., were collected and were embodied in computer programs which were designed to calculate the thermodynamic feasibility of reactions of interest. This program was conducted to obtain a basis on which to establish the thermodynamic feasibility of reactions which the literature survey indicated to be worthy of future study.

The literature survey was conducted throughout the duration of this contract. The other tasks were conducted during the last four months of the contract. All studies were completed between July 1, 1968 and February 24, 1969.

CONCLUSIONS

Based on the literature review and on thermodynamic calculations made from what are believed to be the most reliable available data, the following five approaches are sufficiently promising to warrant conducting laboratory studies to prove their soundness and to develop information needed for further process development:

I. Carbon Regeneration

Present carbon-based processes are severely limited by the requirements of the available regeneration schemes. Thermal regeneration consumes carbon and hyperactivates the sorbent. Regeneration by washing produces weak acid. Regeneration by reduction of the sorbed acid to SO_2 with a gaseous reductant would circumvent the degradation of sorbent while retaining the other advantages of thermal regeneration. Thermodynamically, this approach is possible.

II. Evaluation of Doped Silica Gels

Silica gel, a highly porous matrix which is inert to sulfuric acid, has a modest sorptive capacity for sulfur dioxide. By adding to the gel minor components which are catalytic for the oxidation of SO_2, it should be possible to prepare an adsorbent which may adsorb SO_2 like carbon but which is a regenerable sorbent.

III. Metal Disulfide Sorbents

Nickel disulfide is reported to chemisorb SO_2 at 150°C and to release the SO_2 at somewhat higher temperatures. This will be verified, and other transition metal sulfides, particularly FeS_2 will be tested for similar activity.

IV. Ferric Hydroxide Sorbents

Ferric hydroxide should be investigated in an effort to find forms which are highly active and which will not be deactivated under adsorption process conditions. Its adsorptive capacity for SO_2 is reported to be favorable.'

V. Fly Ash as Lime Carrier

Methods of modifying fly ash to make it a suitable support for hydrated lime in a process similar to the Still process should be studied.

RECOMMENDATIONS

It is recommended that laboratory studies be conducted to obtain the data required for the necessary process engineering analyses in areas where process concepts have been defined. In addition, a laboratory screening program should be instituted to evaluate SO_2 sorption-desorption characteristics of potential new candidate materials for which there are only limited laboratory data. The data from these laboratory studies should be utilized to develop process flow sheets.

ACKNOWLEDGEMENTS

This study was conducted by the FMC Corporation at the Chemical Research and Development Center in Princeton, New Jersey. Mr. R. Bloom, Jr. was Project Manager, Dr. C. A. Gray the Project Scientist, and Dr. L. D. Friedman the Principal Investigator. The literature search was conducted by Dr. H. Perkins. This report was written by L. D. Friedman and C. A. Gray.

SECTION I - INFORMATION SEARCH AND STORAGE

A. PURPOSE

The purpose of the search was to obtain as much information as possible on the removal of sulfur dioxide from gases by inorganic solids other than metal oxides. Since the development of potential systems depended almost entirely on the results obtained from the literature survey, the survey was the first part of the program that was started and it was continued throughout the duration of the contract. The search concentrated principally on adsorption reactions and mechanisms. This approach was followed on the theory that no satisfactory SO_2 removal process could result unless a feasible adsorption process were developed. While this approach places the major emphasis on adsorption studies and mechanisms, references containing material believed to be pertinent to potential or appropriate desorption methods were also taken during the course of this investigation.

B. SOURCES SEARCHED

The major sources searched in conducting the literature survey are listed in Table I. These include Chemical Abstracts from 1907 to date, the Engineering Index, and the Uniterm Index to chemical patents. In addition, general review articles and bibliographies of literature for the past ten years were reviewed to obtain an overall picture of the general background and current status of problems and available solutions. Specialized journals, such as Air Pollution Control Association Abstracts and Fuel Abstracts and Current Titles, etc., were also covered.

Over 1000 references were taken during the literature review. In most cases, the evaluation of the article was made from the abstract of the reference or the patent. However, references which appeared to be of particular interest to this study were ordered, and during the course of this survey about 75 actual papers were obtained for more complete study and an additional 50 or so papers were already available in the FMC library.

C. SCOPE OF THE SEARCH

The literature was searched for references to all materials which are within the scope of this contract and which adsorb or react with SO_2. The materials falling within the scope of this contract include clays, mineral products such as diatomite, industrial wastes, inorganic salts, metals and alloys, covalent compounds, non-metallic elements, oxides of non-metals, and hydroxides.

Both natural and modified clays were considered under the clay subheading. The principal industrial wastes for which references were found were fly ash and slag. Covalent compounds

TABLE I

SOURCES CONSULTED IN THE LITERATURE SEARCH ON SO_2 REMOVAL FROM FLUE GAS

Chemical Abstracts, 1-66(1907-June 1967)

Sulfur dioxide

Chemical Abstracts, Keyword Index, 67(1)-68(26)(July 1967-Dec. 1968)

Absorption
Adsorption
Copper-nickel alloy
Desulfurization
Ferri...
Ferro...
Fly ash
Iron carbide
Iron hydroxide
Iron sulfide
Nickel-copper alloy
Nickel sulfide
Silica gel
Sorption
Sulfur dioxide
Sulfur oxides

The Engineering Index (1931-1968)

- Air pollution
  - (no subheading)
  - Research
- Flue gas
- Sulfur
  - Recovery
- Sulfur compounds
- Sulfur dioxide
- Sulfuric acid

Uniterm Index to (U.S.) Chemical Patents (1952-Oct. 1968)

Sulfur dioxide

Current Chemical Patents (selected for the FMC Corp. Central Research Dept. from the Official U.S. Patent Gazette and Derwent Patent Abstracts for Belgium, France, Germany, Great Britain, the Netherlands, and Japan.)

Vol. 1 nos. 1-4, 6-8, 11, 13 (1966);
Vol. 2 nos. 1-9, 11-20 (1967);
Vol. 3 nos. 2-20 (1968).

Sulfur dioxide removal from flue gas.

TABLE I (cont.)

Air Pollution Control Associations Abstracts

Vol. 14 nos. 1,2(1968); vol. 13 nos. 1-12 (1967);
Vol. 12 nos. 1-9, 11, 12 (1966); vol. 11 nos. 3-5, 7-12(1965).

General aspects
Control methods
Basic sciences - technologies

Fuel Abstracts and Current Titles 3(7) - 7 (12) (July 1962-Dec. 1966).

Sulfur dioxide

British Coal Utilization Research Association Monthly Bulletin 31 (1967), 32 (10) (Oct. 1968).

U.S. CFSTI, SB-448, Rev. Suppl. 1; "Air Pollution and Purification." (Lists reports and translations dealing with air pollution added to the CFSTI collection Sept. 1963 through March 1965).

Bituminous Coal Research, Inc. (J. W. Igoe), "Progress Report on Research on Methods for Control of Sulfur Dioxide Emissions from Coal Burning Power Boilers." Report L-273 (7 March 1968).

Anna Grossman Cooper, Sulfur Oxides and other Sulfur Compounds - A Bibliography with Abstracts, U.S. Dept. of Health, Education, and Welfare, Public Health Service, Div. of Air Pollution, Washington D.C. (1965).

International Chemical Engineering 2 - 8 (1961-1968).

Table of contents

Arthur M. Squires, "Air Pollution: The Control of SO_2 from Power Stacks Part II - The Removal of SO_2 from Stack Gases." Chem. Eng. 74 (24, 133-40 (20 Nov. 1967).

List of 34 references

TABLE I (cont.)

"Proceedings Digest of the 61st Annual Meeting Air Polution Control Association, St. Paul, Minn." (23-27 June 1968).

U. S. Bureau of Mines Bull. 537, "Air Pollution", by S. G. Davenport and G. G. Morgis (1954)1

Recovery of gases and fumes.

included carbide, borides and nitrides. The only non-metallic elements found were silicon and carbon, and virtually all references under this category dealt with carbon. Carbon accounted for more than one-third of all references covered. The only oxide of a non-metal found in the literature was silica, the oxide of silicon.

Oxides of metals fall outside the scope of this contract and were not included in the bibliography. Compounds such as ferrites, molybdates, vanadates, etc. which can be considered either as salts of oxy acids or as mixed metal oxides were included in this survey. The scope of the contract included these substances as adsorbants for removing SO_2 but did not include them as reactants for removing SO_2. This avoided duplicating work being done at Tracor, Inc., under a similar contract where the emphasis is on the chemical reaction between metal oxides and SO_2.

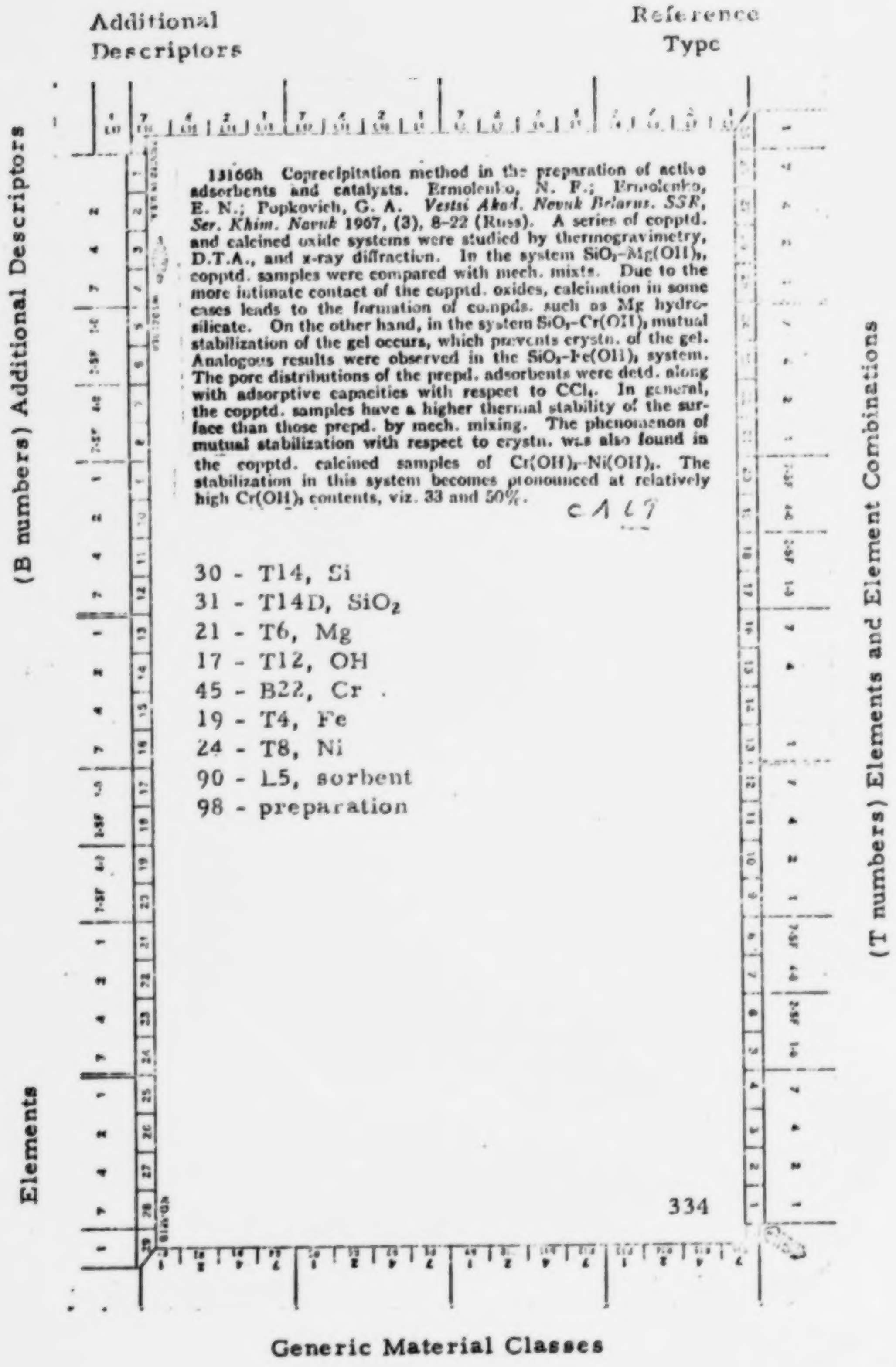
Some references included in the bibliography do not discuss suitable adsorbants or reactants but contain information believed to be useful for devising recovery processes, performing economic evaluations, designing reactors, etc. Other references included were those containing standard thermodynamic values and a representative number of general review articles on methods for solving the SO_2 air pollution problem. References dealing with the catalytic oxidation of SO_2 as used in the contact process for making sulfuric acid usually were not included in this bibliography. However, some of these references were taken and cataloged. A separate file of references dealing with the analytical determination of SO_2 was collected and given to the Analytical Department for possible use in studies to be conducted during Phase II of this contract.

D. REFERENCE CODE SYSTEMS

1. McBee Card Coding System

All references were placed on McBee code cards KD-581B. Where the reference was short or was taken from an abstract publication, the entire reference or abstract was glued to the McBee card. This eliminated the necessity of abstracting references with the possibility of omitting critical data. Longer references were abstracted on the McBee code card. For identification purposes each reference was assigned a unique I.D. number that is indicated on the corner of each McBee card. (See Figure 1)

The information on the McBee cards was coded to include: (1) the nine compound categories that were indicated above; (2) the type of reference; (3) the description of the information; and (4) the element combinations which were discussed in the original article. The code system used is shown in Table II. The reference type indicated whether a reference was a patent, paper, report, or


Additional Descriptors
Reference Type
(B numbers) Additional Descriptors
(T numbers) Elements and Element Combinations
Elements
334
Generic Material Classes

13166h Coprecipitation method in the preparation of active adsorbents and catalysts. Ermolenko, N. F.; Ermolenko, E. N.; Popkovich, G. A. *Vestsi Akad. Navuk Belarus. SSR, Ser. Khim. Navuk* 1967, (3), 8-22 (Russ). A series of copptd. and calcined oxide systems were studied by thermogravimetry, D.T.A., and x-ray diffraction. In the system SiO_2-$Mg(OH)_2$, copptd. samples were compared with mech. mixts. Due to the more intimate contact of the copptd. oxides, calcination in some cases leads to the formation of compds. such as Mg hydrosilicate. On the other hand, in the system SiO_2-$Cr(OH)_3$ mutual stabilization of the gel occurs, which prevents crystn. of the gel. Analogous results were observed in the SiO_2-$Fe(OH)_3$ system. The pore distributions of the prepd. adsorbents were detd. along with adsorptive capacities with respect to CCl_4. In general, the copptd. samples have a higher thermal stability of the surface than those prepd. by mech. mixing. The phenomenon of mutual stabilization with respect to crystn. was also found in the copptd. calcined samples of $Cr(OH)_3$-$Ni(OH)_2$. The stabilization in this system becomes pronounced at relatively high $Cr(OH)_3$ contents, viz. 33 and 50%.

c167

30 - T14, Si
31 - T14D, SiO_2
21 - T6, Mg
17 - T12, OH
45 - B22, Cr
19 - T4, Fe
24 - T8, Ni
90 - L5, sorbent
98 - preparation

Figure 1

Sample McBee Card.

TABLE II

REFERENCE CODE SYSTEM FOR MCBEE CARD PUNCHING

Compound Type	
clays, natural and modified	R0
mineral products (asbestos, diatomite)	R1
industrial wastes (fly ash, slag)	R2
inorganic salts	R4
metals and alloys	R5
covalent compounds (carbides, borides; nitrides)	R6
nonmetallic elements (silicon, carbon)	R7
oxides of nonmetals (silica)	R8
hydroxides	T12

Description of Information	
general review	B5
flue gas	B9
plant operation	B9D
economic discussion	B10
bibliography	L2
sorption	L5
oxidation of SO_2	L6
chemical reaction	L8
desorption	L9
thermodynamics	L10
kinetics	L11

Reference Type	
patent	L1
book	L3
reports	L4
meetings	L4

Elements and Element Combinations			
aluminum	T10	molybdenum	T22
aluminosilicates	T11	nickel	T8
alkali metals (Li, Cs, Rb)	T5D	nitrogen	T27
barium	B24	phosphorus	T24
bismuth	B21	platinum	T18
boron	B25	other platinum metals	T18D
cadmium	B26	potassium	T2
calcium	T28	silicon	T14
carbon	T1	silica	T14D
Reinluft carbon	T1D	fibreglass	T13
carbonate *	T9	bag filterhouse	T13D
chromium	B22	silver	T20
dolomite	T9D	sodium	T5
chlorine	T26	sulfur	T15
cobalt	T23	sulfite or sulfate	T15D
copper	T21	tantalum	B23
gallium, indium, or thalium	T10D	tin	B28
hafnium	T19D	titanium	T17
hydroxides	T12	vanadium	T16
iodine	T25	alkali metal-vanadium compound	T16D
iron	T4	zinc	B27
lead	T3	zirconium	T19
magnesium	T6		
manganese	T7		

* C (8) is not punched for CO_3^{-2} group.

meeting. Headings classifying the description of information included general review, flue gas, plant operation, economic discussion, bibliography, adsorption, oxidation of SO_2, chemical reaction, desorption, thermodynamics and kinetics. Forty-three elements and element combinations were coded on the McBee cards. In addition, special entries were made for Reinluft carbon process, fiberglass, bag filterhouse, and alkali metal-vanadium compounds since these were all mentioned in so many abstracts.

The location of each of these headings on the McBee cards is indicated in Table II. Each designation has a letter and a number. The R and L refer to the R&L numbers on the right and left sides respectively, of the McBee keysort cards. "T" refers to the numbers at the top of the McBee cards, and "B" to the numbers at the bottom. The letter "D" after a number, such as T14D after silica, indicates that this is a deep punch. Thus, the element silicon is T14, but silica is T14D. Silicon references will be included in silica references and would be recovered by the use of the searching needle to separate out those McBee cards containing a punched card for T14. However, the reverse would not be true.

Some rather arbitrary decisions are incorporated in the classification system used. Each potential adsorbant or reactant is classed under the first appropriate heading shown in Table II. Thus, silica is classed under oxides of non-metals but not under covalent compounds. Similarly, clays are not additionally classed under mineral products. A reference can have two material classification numbers if it describes two materials which belong to different material classes, such as references discussing both silica and carbon. A salt such as calcium sulfide is described by the T28 code for calcium plus the T15 code for sulfur. Thus, salts are listed under both anion and cation. Element groups, such as SO_4 and SiO_2 are used instead of the simple element codes for S and Si. However, references coded on the McBee cards contain both element and element group code numbers by using the McBee deep punch. Thus, for example, So_4 is the T15 deep punch for sulfur. Sulfur itself is T15.

Several descriptors may be used to describe the types of information in a reference in addition to those for specific types or compositions. For example, an article which discusses adsorption and desorption of SO_2 on silica is coded under both L5 and L9 which are the codes for adsorption and desorption. If the article also refers to SO_2 in flue gas, then the B9 code for flue gas will also be used. This coding arrangement permits a rather general classification of the various references and at the same time eliminates many references that are not specific for individual headings.

2. Computer Coding System

When it became evident that the number of references was becoming too voluminous to be handled easily by the usual McBee card handling techniques, it was decided to computerize the bibliography. This had the advantage of allowing rapid printouts of selected references, giving rapid and accurate alphabetization, aiding in making specialized searches, and allowing expansion of the coding system that had been used. Consequently, the information on the McBee cards was prepared for a computer system and this information was stored on cards.

Because of the vast amount of time that had been spent coding the references in the McBee cards, it was decided to use a computer coding system which would be based on the classification used on the McBee Keysort card. This procedure had the advantage of using the existing classifications that were immediately available, and while this was not the optimum method to use in computerizing the information, it gave a computerized reference system in the shortest time and with the least expense.

The computer code system finally adopted uses a two digit number to describe each reference type and up to 15 three digit numbers per reference to classify the adsorbent or reactant and to indicate the main points of emphasis in the reference. The use of 15 three digit numbers to qualify a reference was arbitrary. This number could be expanded to any number desired, but it was felt that for our purposes 15 classifiers could adequately describe virtually all the references.

a. Reference Type

The reference type, indicated by a two-digit number, classifies the reference as a journal article, paper presented at a meeting, report, book, article in an irregular publication, a patent, or other type such as private communication. Table III lists the reference types with their corresponding computer and McBee code numbers. Patents are coded for the computer with numbers greater than 10, non-patents with computer-code numbers less than 10. One example of the greater flexibility obtainable with the computerized code was the ability to break the L1 McBee Patent code into eighteen codes to indicate the country of origin of each patent.

Reports under the 01 computer code include publications by Government agencies such as the U.S. Bureau of Mines, or companies, such as Dow Chemical Company. References to papers presented at meetings are classified as such even when the proceedings are issued later in journals or in book form.

TABLE III

REFERENCE TYPE CODES

	Code			Code	
	Computer	McBee		Computer	McBee
journal	00	(none)	Danish	16	(L1)
report	01	(L4)	French	17	(L1)
meeting	02	(L4)	German	18	(L1)
book	03	(L3)	German (E)	19	(L1)
irregular publication	04	(none)	Hungarian	21	(L1)
others	05	(none)	Italian	22	(L1)
Patents					
U.S.	11	(L1)	Japanese	23	(L1)
Australian	12	(L1)	Netherlands	24	(L1)
Belgium	13	(L1)	Russian	25	(L1)
British	20	(L1)	Swedish	26	(L1)
Canadian	14	(L1)	Swiss	27	(L1)
Czech	15	(L1)	Yugoslavian	28	(L1)

b. Material Class

All potential adsorbents and reactants are divided into the nine material classes which were established as the basis of reporting needs of the present contract and given the code numbers shown in Table IV. References which do not discuss a potential adsorbent or reactant have no material class classification. Thus, standard thermodynamic references do not receive such a classification.

Each potential adsorbent or reactant is classed under the first appropriate heading in Table IV. Thus, following the system described above for the McBee card classification, silica is classed under nonmetals but not under covalent compounds, and clays are not included under mineral products.

c. Elements and Element Combinations

The code descriptors listed in Table V were used to assign computer code numbers to elements and elemental compositions. For example, CaS is described by both an 007 (T28) code for Ca and an 036 (T15) code for S. In addition to simple elements, Table V also contains some codes for element groups, such as SO_4 and SiO_2 which are used (if appropriate) instead of the simple element codes for S and Si. Thus, to perform a complete computer search for all references to sulfur compounds one must check references whose code cards contain both 036 (sulfur) and 037 (sulfate). All elements listed under more than one code are listed in Table VI.

The "large number of metals" category is used when the enumeration of all metals listed in a reference would produce more than the 15 allowed three-digit computer code numbers. In these few cases, those elements judged most pertinent were coded individually and the others were indicated by the "large number of metals" code.

d. Additional Descriptors

The descriptors in Table VII indicate the nature of the information contained in a reference. For example, an article which discusses the sorption and desorption of SO_2 on carbon is described by 090 (L5) for sorption and 093 (L9) for desorption. If the SO_2 is in flue gas, then 081 (B0) for flue gas is also used. If the oxidation of SO_2 and its chemical reaction are thought to occur with one substrate both the 091 and the 092 descriptors may be used.

Four terms having no McBee codes were added in the final phase of the search when their inclusion seemed advisable. The processes and systems code includes references to systems definitely outside the scope of the contract, such as the Bureau of Mines' alkalized alumina process.

TABLE IV

COMPUTER CODE FOR GENERIC MATERIAL CLASSES FOR SO_2 REMOVAL FROM FLUE GAS

	Computer Code	McBee Code
clays, natural and modified	070	(R0)
industrial wastes (fly ash, slag)	072	(R2)
mineral products (asbestos, diatomite, natural zeolites)	071	(R1)
oxides of nonmetals (silica)	078	(R8)
hydroxides ($Mg(OH)_2$, FeIII hydroxide)	017	(T12)
nonmetallic elements (carbon)	077	(R7)
metals and alloys (platinum)	075	(R5)
inorganic salts (ferrites, sulfides, synthetic zeolites)	073	(R4)
covalent compounds (carbides)	076	(R6)

TABLE V

COMPUTER CODE FOR ELEMENTS AND ELEMENT COMBINATIONS

	Code Computer McBee		Code Computer McBee
aluminum	001-T10	nitrogen	025-T27
aluminosilicates	002-T11	phosphorus	026-T24
Li, Cs, or Rb	003-B18	platinum	027-T18
barium	004-B24	Ru, Rh, Pd, Os, or Ir	028-B20
bismuth	046-B21	potassium	029-T2
boron	005-B25	silicon	030-T14
cadmium	006-B26	silica	031-T14D
calcium	007-T28	fibreglass	032-T13
carbon	008-T1	silver	034-T20
carbonate	010-T9	sodium	035-T5
chromium	045-B22	strontium	047-none
dolomite	011-T9D	sulfur	036-T15
chlorine	012-T26	sulfite or sulfate	037-T15D
cobalt	013-T23	tantalum	038-B23
copper	014-T21	thiocyanate	050-none
gallium, indium, or thalium	015-B19	tin	039-B28
iodine	018-T25	titanium	040-T17
iron	019-T4	tungsten	048-none
lead	020-T3	vanadium	041-T16
magnesium	021-T6	alkali metal-vanadium compound	042-T16D
manganese	022-T7		
molybdenum	023-T22	zinc	043-B27
nickel	024-T8	zirconium	044-T19
		large no. of metals	050-none

TABLE VI

ELEMENTS INCLUDED UNDER MORE THAN ONE COMPUTER CODE LISTING

Element	Code Numbers
aluminum	001 or 002
Li, Cs, or Rb	003 or 042
calcium	007 or 011
carbon	008 or 010 or 011 or 050
magnesium	021 or 011
nitrogen	025 or 050
potassium	029 or 042
silicon	030 or 031 or 032 or 002
sodium	035 or 042
sulfur	036 or 037
vanadium	041 or 042

TABLE VII

ADDITIONAL COMPUTER DESCRIPTORS

general review	080 (B5D)	sorbent preparation	098 (none)
flue gas*	081 (B9)	sorption	090 (L5)
plant operation+	082 (B9D)	oxidation of SO_2	091 (L6)
engineering	099 (none)	chemical reaction	092 (L8)
processes and systems	097 (none)	desorption	093 (L9)
Reinluft process	009 (T1D)	thermodynamic data	094 (L10)
bag filterhouse	033 (T13D)	kinetics	095 (L11)
economic evaluation	083 (B10)	recovery	096 (none)
literature survey	085 (L2)		

* Compositions like that from power plant stacks and not smelter gases or tail gases from manufacturing sulfuric acid.

+ Includes description of pilot plant operations.

Because it is mentioned so often, the Reinluft process has a separate code. Recovery can refer to either recovering the sulfur value in the flue gas or generating the spent adsorbent or reactant. Engineering is used to indicate information such as gas-solid contacter designs.

3. Computer Input

The computer input for each reference consists of from one to eight cards (usually 2 or 3) which contain the citation plus one card which contains all numeric code information. The cards containing the citation are numbered sequentially (starting with 1) in column 80. The code card (also called the 9-card because it contains a 9 in column 80) is the last card for a given reference and contains all the code numbers. All cards belonging to one reference contain its identifying (I.D.) number in columns 76-79 for non-patents or in columns 75-79 for patents. A variable number of blank columns occur prior to the ID number since it is undesirable to have words split arbitrarily. The coding system is discussed more fully below.

a. Citation Input

The format used for keypunching the citation has been standardized to allow computer searching for authors, title, journal, patent number, and year. These five items have specific tags by which the computer can identify them.

(1) Author Information

Author information is always at the beginning of the citation and includes individuals, companies or government agencies issuing reports, assignors of patents, and inventors. Several examples follow:

1 *Smith I M, Taylor R P, Jones H K, et al.*
2 *Dow Chem Co* (Smith I M, Taylor R P, Jones H K, et al).
3 *Dow Chem Co*
4 *U S Bur Mines* (Smith I M).
5 *Smith I M, Taylor R P*

The author information for a journal article (or an unassigned patent) is keypunched as in example 1 for more than three authors (inventors) or as in example 5 for two authors (inventors). The author information for an assigned patent with inventors Smith, Taylor, Jones, et al. is keypunched as in example 2, or, if the inventors are not known, as in example 3. Reports are keypunched as in example 4 (Smith is the author) or example 2 (Smith, Taylor, Jones, et al. are the authors and Dow Chemical Co. is issuing it). The first asterisk occurs in column 1 of card 1 and the first period always occurs at the end of the author information. This period is sometimes followed by an asterisk.

(2) Title

The title is always the second unit of a citation. It is immediately preceeded and ended by an 028 multipunch. The 028 multipunch indicating the beginning of the title sometimes occurs directly after the period-asterisk ending the author information, sometimes directly after the period ending the author information, sometimes one blank column removed from the period ending the author information, or in rare cases (when the author information uses the whole first card) in column 1 of a new card. The KWIC mark prints out as a dash (see Keypunched sample in Table VIII).

(3) Journal

The journal name is the third piece of information for journal references. Its name is immediately preceeded and followed by the slash symbol, the /. This slash[1] occurs immediately after the second 028 KWIC mark, or in column 1 of a new card when appropriate. Periods are used after abbreviations in the journal names. Because Chemical Abstracts has changed its abbreviations several times over the past fifty years and also because references were obtained from other sources which use different abbreviations, a computer search was performed for journal names and an alphabetical listing produced. This ensured that all references to a given journal had an identical abbreviation. Slashes are also used to indicate location of other non-patent references, such as meetings, conferences, etc.

(4) Year

The year usually is placed in parenthesis at the end of a reference. However, if a journal has no volume number the year will be placed immediately after the name of the journal. When the year is at the end of a journal reference, it may be accompanied by either the day and month or by the month alone. Thus, referring to Table VIII, reference number 0065 is dated August, 1967, whereas reference 1008 is dated 20 Dec. 1967. Patent dates always contain the day, month, and year. The < symbol is always placed before and after the year so that there can be no confusion as to which number constitutes the year of the reference. Commas, parenthesis, and periods may also be included. Examples include (27 June < 1968). <,<1968<, (1968). The < symbol prints on the reference sheet as a square (see Table VIII).

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[1] The slash is also used to indicate patent numbers.

TABLE VIII

SAMPLE REFERENCE SHEET AS KEYPUNCHED

```
FEBRUARY 24, 1969                                                                                PAGE  1

   1      *DRATWA H, JUENTGEN K, PETERS W.*-MECHANISM OF FORMATION OF SULFUR DIOXIDE 00651
   2      ON ADSORPTION COKE USED IN DRY WASTE GAS PURIFICATION.-/CHEM. INGR. TECH./ 00652
   3      39 (16), 949-55 (AUG□1967).□                                               00653
   4      00008090091093077                                                          00659
   5      *GAUBE J, KNACKE O, LYDTIN H, ET AL.*-METALLURIGICAL PHASE DIAGRAMS OF     00931
   6      IRON COMPOUNDS.-/ARCH. EISENHUETTENW./34 (9), 653-8□(1963).□               00932
   7      00019036037094092073                                                       00939
   8      *DRATWA H, JUENTGEN H.*-DESULFURIZATION OF SMOKE GASES WITH ABSORBENT      03551
   9      CHARCOALS WITH DIFFERENT PROPERTIES.-/STAUB, REINHALTUNG LUFT/27 (7), 301-703552
  10      □(1967).□                                                                  03553
  11      00008081090095093077                                                       03559
  12      *U S NATL AERONAUTICS AND SPACE ADMIN*(MCBRIDE B J, HEIMEL S, EHLERS J G,  05181
  13      ET AL).-THERMODYNAMIC PROPERTIES TO 6000 DEG K FOR 210 SUBSTANCES          05182
  14      INVOLVING THE FIRST 18 ELEMENTS.-SP-3001, WASHINGTON, D.C.□(1963).□        05183
  15      01094                                                                      05189
  16      *U S BUR MINES*(KELLEY K K).-CONTRIBUTIONS TO THE DATA ON THEORETICAL      05151
  17      METALLURGY. VII. THE THERMODYNAMIC PROPERTIES OF SULFUR AND ITS            05152
  18      INORGANIC COMPOUNDS.-/BULL./406□(1936).□REPRINTED IN BULL. 601 (1960).     05153
  19      01094                                                                      05159
  20      *TRACOR CORPORATION.*-THERMODYNAMIC PROPERTIES OF SELECTED METAL           05111
  21      COMPOUNDS.-/PRIVATE COMMUNICATION/(11 NOV□1968).□                          05112
  22      05094                                                                      05119
  23      *DOW CHEMICAL CO*(STULL D R).-JANAF INTERIM THERMOCHEMICAL TABLES.-        05261
  24      MIDLAND, MICH.□(1960)□ET SEQ.                                              05262
  25      01094                                                                      05269
  26      *BILLINGE B H M.*-THE CHEMISORPTION OF SULFUR DIOXIDE ON CARBONS.-/CONF.   04291
  27      IND. CARBON GRAPHITE, PAPERS, 2ND, LONDON/1965, 398-404 (PUB.□1966).□      04292
  28      02090008077093                                                             04299
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(5) Patent Number

In classifying patents, the third piece of keypunch information is the name of the country of origin, abbreviated with no period, followed by a slash (/), the patent number, another slash (/) and then the date. For example, Brit/1,054,798/ (27 Nov <1965).< Note that a search for a patent number requires searching first for the reference type code corresponding to the desired country followed by a search for the patent number.

b. Code Input

The code card accompanying each citation contains the two-digit code number for reference type in columns 1 and 2, and up to the maximum of fifteen three-digit code numbers in columns 3-48. The three-digit code numbers can be arranged in any order.

4. Function of Computer Programs

Listing Program - 1

This program lists the file of cards punched according to the coding system described in IV above in the general format used in this report (format 1). Specifically, this program

(1) deletes *,/,<, and the 028 multipunch from the print out

(2) deletes the I.D. code number from all cards except the code card

(3) deletes the digit punched in column 80. This is the card number for each reference when more than one card per reference is used.

(4) corrects the left hand margin of the 1st card of a reference for the deleted*

(5) inserts one space between each code number listed on the 9-card

(6) inserts a blank line between the 9-card of one reference and the beginning of the next reference.

Listing Program - 2

This program lists the references in format-1 except that it numbers the references in sequential order. This order may be alphabetical by author, by codes, or by the I.D. number.

Code-Search Program - 1

This program can print out in format-1 references whose 9-card contains a given three-digit code number. Since the appropriate references are listed in the same order as they occur in the file, the alphabetical file is used for making alphabetical listings.

Code-Search Program - 2

This program lists the reference I.D. numbers of all references which do not contain a three-digit code number of the form 07X where X can be 0, 1, 2, 3, 4, 5, 6, 7, or 8. This program was used to check that all appropriate references had a material class code number.

I.D. Search Program - 1

This program lists in format-1 references corresponding to the indicated I.D. numbers. An alphabetical listing is produced when applied to the alphabetically arranged deck of cards.

Bibliographic-Information Search Program - 1

This program produces a deck of punched cards (one per reference) which duplicates the information (journal name) between the two slashes (/) provided it occupies no more than 76 columns. The last four columns of the punched card contain the reference number. This is useful for collating all references from a given journal (or source). If this deck of punched cards is alphabetized by an 082 sorter and printed using a standard 80/80 listing, an alphabetical listing of the journal names is obtained.

SECTION II - EVALUATION AND DISCUSSION OF THE RESULTS OF THE LITERATURE SURVEY

During the course of this study, over 1000 references and patents were evaluated critically. About half were rejected because they were not considered pertinent to the project. Most of the rejected references dealt with the use of platinum and vanadium oxides materials to oxidize SO_2 to SO_3 for the manufacture of sulfuric acid.

Our principal interest in this study was in determining the state of the art for removing SO_2 from flue gases containing only about 0.35 mole percent SO_2. Unfortunately, most references that discussed the adsorptive behavior of or the reaction of SO_2 described the behavior of streams of essentially pure SO_2. Thus, many of the reactions which involve SO_2 would not remove low concentrations of SO_2 from flue gases. Whenever thermodynamic data were available, the results were extrapolated from the SO_2 concentration reported to the concentration in flue gas.

All references were coded and grouped according to the generic material classes, and each class was studied separately. In some cases, the generic classes were broken down further. Thus, because of the large number of iron references collected, the iron salts were reviewed by themselves as a sub-group of the inorganic salts class. The iron references included a few references from classes other than the inorganic solids, such as iron-doped silica gel, iron slags, etc.

From the review of the literature, those references which appeared to be most interesting based on the degree of reactivity reported with SO_2, or the quality and quantity of work done, or the thermodynamic or other data listed, were collected. In a few cases where available thermodynamic data were located, an attempt was made to evaluate the thermodynamic potentialities of the reactions described. The following nine sections describe what are believed to be the most pertinent references located for each of the nine classes into which our general category was broken.

A. Clays

The only clay mentioned as being an adsorbent for SO_2 is bentonite (245,429). A fairly recent reference claims that oxides of sulfur are adsorbed strongly on acid-washed bentonite.

The heat of adsorption of water on activated bentonite is two to three times that of the oxides of sulfur. (429) Because the moisture content of flue gases is many times that of SO_2, it is assumed that clays probably would adsorb moisture rather than the SO_2. However, because of bentonite's relatively low cost and superior resistance to oxides of sulfur, bentonite may be evaluated briefly with synthetic flue gas as part of our overall screening technique development program in Phase II to verify our assumption.

B. Mineral Products

Many investigators and organizations currently are studying the use of lime and dolomite to remove sulfur dioxide and other oxides of sulfur from flue gases. The processes under study are direct injection of lime into furnaces and flue gases followed by scrubbing the waste gases with aqueous suspensions of lime. Lime treatment has been discussed in great detail in many review and specialized articles. Because of the wide-spread interest in the use of lime for removing oxides of sulfur, it is felt that no further studies with lime should be undertaken within the scope of this contract.

U.S. patent, 3,286,106 describes the reaction of oxides of sulfur on dry chrysotile asbestos $Mg_3Si_2O_5(OH)_4$. It is claimed that asbestos can combine with from 0.01 to 3.0 weight percent sulfur and that the sulfurized asbestos is a superior filler for vinyl tile. Asbestos would have excellent inertness to SO_2 but its low adsorptive capacity would rule out consideration of its use as an adsorbent for oxides of sulfur.

The possibility of forming fertilizer by reacting phosphate rock with SO_2 gas has been studied (373,161). These studies showed that a slurry of phosphate rock adsorbed enough SO_2 to make the phosphate soluble. This process was considered as a possible means for making low cost fertilizers, but was abandoned when it was found to have no economic advantage over the use of concentrated sulfuric acid. Dry treatment of a phosphate rock with sulfur dioxide at temperatures above 95°C rendered about two-thirds of the P_2O_5 available as a fertilizer. Part of the SO_2 was decomposed to sulfur and some formed calcium sulfate and calcium pyrosulfate.

The investigators who studied the reaction of SO_2 with phosphate rock hoped to use concentrated SO_2 gas rather than sulfuric acid to manufacture fertilizer. Even though this reaction did not appear promising, it is possible that phosphate rock could adsorb substantial quantities of SO_2 from flue gases and that the spent sulfated phosphate rock might then be used as feed to a fertilizer plant. Since phosphate rock occurs in many parts of this country and since the reaction with SO_2 occurs at temperatures above the dew point of the two gases, it might be desirable to examine the reaction of SO_2 with phosphate rock to determine whether enough SO_2 could be adsorbed from flue gases to justify the handling costs and to reduce the amount of sulfuric acid required for fertilizer manufacture.

C. Industrial Waste

Several industrial wastes have been examined to determine their ability for removing oxides of sulfur from flue gases. Two recent patents claim the use of fly ash for the removal of SO_2 but give little data on the type of fly ash used or the sorption conditions studied (517, 473). Iwamura (166) claims that copper blast furnace slag has good SO_2 sorption properties. A study of the translation of this paper casts considerable doubt as to the usefulness of such slag for removing SO_2 from flue gas.

Billings (39) discusses the removal of oxides of sulfur by the use of mineral wool filters. Filters two inches thick and with a density of four pounds per cubic foot removed 80% of acid mist and 99% of acid gas and fumes. By washing away the iron oxide and fly ash which became trapped in the filters, he was able to reuse the filters 8 to 10 times with no apparent loss of SO_2 removal capacity. The removal of acid gas was improved by the presence of moisture on the slag wool filters. Mineral wool filters, according to Billings, have the advantage of low cost, small fiber diameter, the ability to withstand temperatures of 1000°F, and the ability to remove submicron particles from industrial furnace effluents. Pressure drops across the filters were not stated. Because these filters remove acid gases and mist so effectively, Billings suggested a two stage process in which flue gas is oxidized at high temperatures to convert the oxides of sulfur to SO_3 (using potassium oxide and vanadia catalysts to effect the oxidation), and the acid mist is removed on the mineral wool filter at temperatures just above 125°C. He claims that this temperature is low enough to achieve good SO_2 absorption but high enough to make reheating of the gas unnecessary.

Red mud, a by-product of aluminum manufacturing which has a relatively high iron oxide content, removed 70 to 75% of the SO_2 in a flue gas, according to a communication from the Republic Coal and Coke Company. The process is being investigated in pilot plants, but no commercialization has yet been attempted. Red mud has only limited availability. Its properties vary from location to location and generalization of its reactivity with SO_2 is difficult.

The Still process uses hydrated basic lignite ash to adsorb SO_2 (350. This process is described in considerable detail on pages 126-28. The lignite ash may or may not be augmented or supplanted with hydrated lime. The process operates in the temperature range between 200 to 850°F. Oxygen and carbon dioxide do not interfere with the operation of the process, according to the description of the process in the literature. The reaction occurs in a transport bed and is primarily the reaction of calcium hydroxide with SO_2. Thermochemical data indicate that the reaction is highly favorable and that the temperature of operation is limited only by the decomposition of the calcium hydroxide, which is above 580°C. Regeneration is somewhat difficult because part of the calcium sulfite that forms is oxidized to calcium sulfate which can not be regenerated. Calcium sulfite can be decomposed thermally to calcium oxide, which can be reused, and to SO_2, which can be recovered. The adsorbent is inexpensive enough so that partial regeneration may be practiced or the spent adsorbant may be discarded entirely. A somewhat similar process was described by Herzog (153), who combined SO_2 with the basic constituent of ash. His SO_2 removal reaction is also based on the reaction of calcium oxide with SO_2, and according to the author is a promising commercial development in East Germany.

The use of a fly ash to remove sulfur dioxide has considerable merit. Many fly ashes are basic and their natural basicity could be used to remove sulfur dioxide. In addition, it should be possible to modify fly ash to make it a suitable support for hydrated lime which would react with the oxides of sulfur. Such a preparation would not be too different from that used in the Still process, but it would be more generally available than the lignite ash which the latter process uses. The difficulties encountered in regenerating the adsorbant from the Still process would not be a factor in the use of alkalized fly ash since the wasting of a considerable amount of the spent fly ash would be of slight economic importance.

D. Inorganic Salts

Over 200 references have been located which mention the use of inorganic salts as adsorbers for SO_2. In many cases, too little information was presented to permit a full understanding of the processes involved and the papers were ordered. Several of the reactions occur in high partial pressures of SO_2, but would not be applicable for flue gas cleanup, based on simple thermodynamic calculations. In other cases, such data were not available and the reaction could not be evaluated properly.

It would be expected that those elements which are mentioned most often in the literature as being adsorbents for or reactants with sulfur dioxide, would be the ones that would be the most active and would be the element around which potential processes should be considered. On this basis, the elements mentioned most often in the literature are the alkaline metals of Group 1, the alkaline earth metals of Group 2, and the transition metals of Group 8 of the Periodic Table. Other elements mentioned less often include aluminum, indium, tin, lead, vanadium, chromium, manganese, and tungsten. The salts of these metals that are mentioned most often include sulfides, sulfites, sulfates, halides, carbonates, and ferrites. Two of the better known sulfur dioxides removal processes use combinations of these elements. These are the Grillo process and the sodium aluminate process.

Nickel disulfide has some interesting properties for reacting with SO_2. Nickel disulfide chemisorbs SO_2 at temperatures of about 120°C, according to Budelot and Delafosse (54). The article was concerned primarily with a sorption phenomena at these temperatures and does little more than point out that chemisorption exists, and appears to increase with increasing temperature. Interestingly, nickel disulfide also catalyzes the reduction of SO_2 at higher temperatures so that a potential process, based on a chemisorption at 120°C, and desorption at temperatures above 200°C, would appear to be possible.

One problem in working with nickel disulfide is the fact that the literature mentioned many nickel sulfide compounds. A recent Russian article discussing the steady-state potentials of nickel sulfides points out that samples containing from 21.1 to 35.44% sulfur showed markedly different chemical, microstructural and surface properties.

An interesting thermodynamic analysis of the reduction of sulfur dioxide by metal sulfides with the formation of elemental sulfur was prepared by some Russian investigators (213). In the case of sulfides of alkali and alkaline earth metals, these investigators claim the equilibrium yield of sulfur at 500 to 800°C was determined to reach 95+ weight percent with on other by-product gases being formed. By reacting with sulfur dioxide and oxygen in the flue gas, the sulfides were converted to sulfates which can be reconverted to sulfides by the action of a reducing gas. According to these investigators, the equilibrium yields of the reduction of sulfates to sulfides by carbon monoxide in the temperature range of from 800 to 1100°C is over 90%.

Wohler, et al., describe the reaction of SO_2 with calcium, iron, and zinc sulfides and with many ores, including pyrites (434). At first glance, it would appear that iron sulfides should react similarily to nickel sulfide and that this reaction was not unusual. However, a thermodynamic investigation showed that the reaction of iron sulfide with SO_2 occurs only at relatively low temperatures and probably at higher SO_2 partial pressures than would be obtained from flue gas. British patent 726,216 claims the formation of Fe_3O_4 and sulfur when a stream of oxygen and SO_2 are passed over ferrous sulfide at 900 to 1000°C.

Sodium silicates have been reported to adsorb as much 40 weight percent of SO_2 at temperatures of 300 to 550°C. This reaction is discussed in greater detail in the section on silica below.

Several investigators mention the use of barium salts for the adsorption of SO_2 over the temperature range of from 100 to 400°C (165, 565, 548, 67). One reference claims that the sorptive action of barium carbonate was improved when the barium carbonate was mixed with 20 to 43% of carbon (67). The function of this carbon is not clear, since the abstract gives no indication of the formation of carbonates or carbides.

Erdos analyzed the properties of systems consisting of gaseous SO_2, CO_2, and a solid metal oxide with respect to the equilibrium pressures of SO_2 (106, 107, 108, 109). In discussing 24 systems for removing SO_2 from flue gases, Erdoes considers the CO_2 content of the flue gas to be sufficient to convert many of the metal oxides to metal carbonates at temperatures below 250°C. Thus, although he starts with metal oxides, he is really discussing the adsorption of SO_2 on carbonates in many cases. The reaction rate of sodium carbonate with SO_2 below 150°C is quite dependent on the water vapor content of the SO_2-air mixture. This was confirmed by Russian workers (195).

The reaction of SO_2 with phosphate rock was reported above. It is interesting to note that the literature contain almost no references to the use of phosphates for SO_2 removal. The reason for this apparent anomaly is not clear.

The survey of the adsorption of SO_2 by inorganic salts suggests that some sulfides, particularly nickel sulfides, are worth studying seriously as potential adsorbents for sulfur dioxide. Other transition metal disulfides, particularly FeS_2 should also be tested for similar activities. We plan to evaluate the sorptive capacities or abilities of many compounds using relatively short reaction periods under Phase II and will check out many of the other compounds that have been mentioned repeatedly in the literature to observe their reactivity with or sorption of SO_2 in concentrations approaching those found in flue gases.

E. Metals and Alloys

A large number of investigators have studied the adsorption of SO_2 on metals and various alloys. Platinum on silica is mentioned many times in connection with the conversion of SO_2 to SO_3 in the manufacture of sulfuric acid. Pevnyi (280) claims that at 450°C, chromium-tin alloys activated by antimony have better adsorption characteristics than either platinum or asbestos or platinum on silica gel. Barium-vanadium and barium-aluminum-vanadium alloys were less active. The difference in sorption activities between all these different alloys and metals disappear at 500°C. Danilova (70) reports moderate reaction of SO_2 with zinc, cadimum, copper, and lead.

Pannetier has done extensive work on the reactions of SO_2 with nickel, iron, and cobalt (265, 266, 267, 268). He claims that the reaction of SO_2 with finely divided nickel is very exothermic and proceeds rapidly at 460°C according to the equation: $7Ni+2SO_2=4NiO+Ni_3S_2$. Thermogravimetric and x-ray studies showed that SO_2 reacts with Ni_3S_2 at 460-550°C to form NiO and NiS, and the latter is finally transformed into NiO with more SO_2. The advance of the interface is the limiting factor and the energy of activation is 84 kcal/mole.

Rumyantsev (307) agrees that nickel reacts with SO_2 according to the equation shown above. He reports that a vigorous chemical reaction starts at between 460 and 470°C but that the reaction is most nearly complete at between 600 and 800°C. He also claims that reverse reaction is possible at temperatures above 800°C.

Perhaps part of the difficulty is interpreting the data from the reaction of nickel and SO_2 arises from the fact that one of the products, nickel sulfide, also reacts with SO_2 to form the oxide and sulfur. (See Section 4). However, the latter reaction occurs at lower temperatures than those used by Pannetier and Rumyantsev. In the temperature ranges they used, the reverse reaction of nickel oxide with sulfur is favored.

Other metals reported to react with SO_2 include indium (263), rhodium (423), copper (314), magnesium (321), and alkali metal sulfate-vanadium oxide mixtures (372). The reaction between cobalt, nickel, iron, and indium with SO_2 is probably a chemisorption reaction since the metal is usually converted to the sulfide and oxide. With nickel and cobalt, the reaction is reversible at temperatures above 800°C. Indium requires higher temperatures to give a reversible reaction.

Platinum is known to be an excellent adsorbent for SO_2 but is not being considered because of its cost. However, a nickel-copper alloy which is reputed to have the same electron configuration as platinum is known. If this material can be prepared its SO_2 adsorption behavior will be checked.

F. Covalent Compounds

Our classification for covalent compounds includes carbides, nitrides, and silicides. No nitrides, or silicides were cited as being adsorbents for SO_2. It has been reported that silicon carbides, or carborundum as it is better known, adsorbed a negligible amount of SO_2. Nickel carbide adsorbs traces of SO_2 at ambient temperatures.

In the temperature range of 250 to 400°C, nickel carbide reacts with SO_2 to form mainly CO_2 and NiO. Small amounts of Ni, Co, Ni_3S_2 and sulfur also form.

G. Non-Metallic Elements

The only non-metallic element which adsorbs appreciable amounts of SO_2 is carbon. At least three of the processes described below as having been studied in commercial or semi-commercial plants are based on the use of carbon as an adsorbent. As a result of this interest in the commercialization of the use of carbon for SO_2 removal, a vast amount of work has been done studying the reactions of SO_2 on or with carbon. Thus, approximately one-third of all references obtained to date, deal in some way with the reactions of SO_2 with carbon.

The Reinluft G.m.b.H. has several patents describing the use of a carbonaceous adsorbent for removing SO_2 from gases. (534, 535, 536, 537, 538). They suggest the introduction of ammonia into the carbon to aid reaction with the sulfur dioxide. This is only one of a series of ideas in patents which have been granted to various companies to improve the adsorption characteristics of carbon. Other materials that have been added to or have been impregnated on the carbon, include alkaline metal and ammonium iodides or iodates, iodine itself, and various promoters, such as manganese, copper, magnesium, iron, zinc, nickel, cobalt, chromium, vanadium, molybdenum, tin, and their respective oxides (U.S. Patent 3,318,662). It has been reported also that activated carbon impregnated with an alkaline material has better SO_2 adsorption characteristics if it is also impregnated with a humectant. (U.S. Pat. 3,396,122). This would suggest that moisture improves the adsorptivity of carbon but liquid humectants, such as the glycol or polyvinyl alcohol specified in the reference cited, would probably evaporate or be swept off the carbon by the high temperature flue gas.

J. Siedlewski has investigated in great detail the effect of the properties of activated carbons on their use as an adsorbent for SO_2 (329 thru 345). He indicated that free radicals have no influence on the physical adsorption of SO_2 but are active centers for its chemisorption. He claimed that adsorbed H_2S prevents the contact of free carbon radicals with oxygen. From studies on the role of free carbon radicals in the oxidation of SO_2 to SO_3, he found a correlation between chemisorption and free radical concentration. This reaction is promoted by carbon. Somewhat surprisingly, Siedlewski reported that covering the surface of carbon with sulfur did not reduce the possibility of oxidation of SO_2 to SO_3. Instead, the reverse might actually occur.

The thermodynamics of various reactions by which sulfuric acid and SO_3 may be removed from carbon, were checked by the computer and the potential carbon regeneration reactions having a favorable Log K are shown in Table IX. Reaction 3 is believed to be the main reaction that occurs during regeneration of the Reinluft process and the reaction most responsible for the degradation of the carbon. Reaction 4, although thermodynamically favorable, is reported to occur to a much smaller extent. It should be noted that of the seven reactions listed in Table IX, four employing hydrogen or CO as a reductant have favorable thermodynamic properties. This reinforces the belief that it should be possible to regenerate carbons that have adsorbed SO_2 without having the carbon react with either SO_2 or sulfuric acid that forms on the carbon surface or in its pores. The action of SO_2 and carbon is discussed in greater detail on page 42 of this report.

TABLE IX

CARBON REGENERATION REACTIONS
HAVING FAVORABLE LOG K

$H_2SO_4 + CO = H_2O + SO_2 + CO_2$ (1)

$H_2SO_4 + H_2 = 2H_2O + SO_2$ (2)

$2H_2SO_4 + C = 2H_2O + 2SO_2 + CO_2$ (3)

$H_2SO_4 + C = H_2O + SO_2 + CO$ (4)

$SO_3 + C = SO_2 + CO$ (5)

$SO_3 + CO = SO_2 + CO_2$ (6)

$SO_3 + H_2 = SO_2 + H_2O$ (7)

The most recent information indicates that the Reinluft process has been abandoned because the carbon used to adsorb the SO_2 became pyrophoric after repeated adsorption-desorption cycles. The sorption of SO_2 on active carbon is not a simple adsorption. Instead, the SO_2 is probably oxidized by the surface oxides to SO_3 and the SO_3 reacts with water present in flue gas to form sulfuric acid which collects in the pores of the carbon. This sulfuric acid cannot be removed thermally without causing a considerable amount of reaction between the sulfuric acid and the carbon. The result is that the carbon surface area gradually increases during repeated regenerations and the carbon becomes pyrophoric. The pyrophoricity arises from the fact that the residual carbon skeleton is too small to conduct the heat of adsorption from the surface, and as a result, a build up of heat occurs within the carbon bed, leading to burning of the carbon.

Carbon is basically an ideal adsorbent for SO_2. It adsorbs SO_2 from dilute streams, it is inert to SO_2, SO_3 and sulfuric acid at ambient temperatures and it can be prepared at relatively low cost. Since the main objection to the use of carbon is in the regeneration which leads to its destruction, it has been suggested that work be done to study the regeneration of carbon. This work will involve trying to make use of those reactions which were shown in Table IX to remove the sulfuric acid without attacking the carbon and leading to its ultimate destruction.

H. Oxides of Non-Metals

The only oxide of a non-metal having significant SO_2 adsorption properties is silica. Silica gel has been studied extensively as an adsorbant for SO_2 because of two very desirable properties. It adsorbs SO_2 reversibly and is not affected by any sulfuric acid that accumulates in the pores. Adsorption of SO_2 on silica gel is aided by increased surface area of the gel (239, 241, 270). One drawback to the use of silica gel as an SO_2 adsorbent is the fact that it adsorbs water as well as sulfur dioxide; and since the concentration of water in flue gas is many times that of sulfur dioxide, silica gel probably would not remove SO_2 from flue gas effectively. However, those silica gels which have the greatest capacity for adsorbing SO_2 usually contain a minimum of about 5% of adsorbed water (270).

A fairly recent paper by Jones and Ross (174) presents data that can be used to estimate the amount of SO_2 that could be adsorbed from a flue gas. Their data show silica gel to be a poor adsorbant for SO_2 under flue gas conditions. These investigators plotted the weight of SO_2 adsorbed per gram of silica gel vs. the partial pressure of SO_2 at 323°K, the highest temperature studied. According to their data, the amount of SO_2 adsorbed from a gas having an SO_2 concentration similar to the 300 ppm concentration found in flue gas is estimated to be only about 6 mg per g. To extrapolate these data to the point where 90% of the SO_2 from flue gas would be removed at 323°K, which is too low for flue gas treatment, 1 gram of silica gel would be needed for every six-tenths mg of SO_2 removed. They also found that heat of adsorption of SO_2 on silica gel varied from 9.3 Kcal. on fresh silica gel to 7.8 Kcal. per mole on silica gel that had a high degree of surface coverage with SO_2. Thus, the heat of adsorption of SO_2 on silica gel is about twice the heat of condensation of SO_2, which is 5.1 Kcal. per mole. This indicates that adsorption of SO_2 on silica gel involves something more than merely a phase change.

Although silica gel itself is not a promising adsorbent for SO_2, many references claim that doped silica gel adsorbs fairly large concentrations of SO_2. Thus, sodium oxides, vanadium, vanadium plus tin, and many other combinations are reported to enhance the adsorption of sulfur dioxide on silica gel. Takats (369) studied the adsorption of SO_2 from SO_2-air mixtures using doped silica gel containing 1, 2, 3, and 4 moles of silica (SiO_2)

per mole of sodium oxide (Na_2O). He showed that the most active compound was $Na_2O \cdot 2SiO_2$, and that the weight percent of SO_2 adsorbed on the sodium silicate calculated as SO_4 reached 60.9% at 550°C for the $Na_2O \cdot 2SiO_2$, compound. The sodium oxide silica compound containing a 1:1 molar ratio was the next most active. Hodek (155) reported excellent adsorption on the use of vanadium-doped silica gel. Vidra (421) prepared a series of silica gel containing ferric ions and claimed that the preparations were complicated and depended on the buffer composition. Levy (225) reported on the interaction of SO_2 with iron on silica bases, including silica-alumina. He claimed the formation of both sulfate and sulfite ions. Pevnyi (280) studied the adsorption and oxidation of SO_2 on silica gels activated with chromium and tin, platinum, and with barium-vanadium compound. All adsorbed far more SO_2 than undoped silica gel. Dzis'ko (197) described the preparation of many silica gels in detail.

A Russian paper (110) describes the preparation of active silica gel adsorbants by coprecipitation followed by calcination. Their results showed silicate formation in the SiO_2-$Mg(OH)_2$ system but gel formations in the SiO_2-$Cr(OH)_3$ and SiO_2-$Fe(OH)_3$ systems. The formation of gels in these systems was attributed to mutual stabilization caused by the respective ions which prevented crystallization of the gels. Co-precipitated samples had higher thermal stability of the surfaces than those prepared by mechanical mixing.

A Netherlands patent application (551) claims a process for using metal-doped silicas to adsorb SO_2 from oxygen-containing gases. Metal doped silica-alumina and silica-magnesia are also mentioned. The metals used to dope the carriers were 5 to 15 weight percent copper, 0.1 to 10 weight percent chromium, and 0.1 to 2 weight percent barium oxide. The patent claims that SO_2 is adsorbed at 325 to 425°C, and is removed at from 350 to 450°C by passing a reducing gas mixture containing hydrogen, carbon monoxide, methane, ethane, propane, or butane through the catalyst bed. Other metals mentioned as possible impregnants include alkali metals, and iron. These metals can also be regenerated by the use of a reducing gas.

The ability of silica and doped silica gels to remove SO_2 and SO_3 from gas streams is apparently a function of gel porosity., According to Chufarov (43) the adsorption of SO_2 increased as the porosity of the gel increased, until a maximum was reached. Then greater porosities did not lead to greater further adsorption of SO_2, presumably because of poor diffusion of SO_2 into the gel structure. Kharmandar'yan (198) state that the sorptive capacity of silica gel for SO_2 did not depend on total pore volume but rather on the differential distribution of the pore volume, and claimed that 7.65 A radius was the most effective pore size. He also reported that iron salts gave more active silica gel than salts of aluminum and manganese. McGavack (239) studied different methods of preparing silica gels and how their preparations affected the adsorptive capacities at temperatures of from -80 to 100°C. He reported that gels containing from 4.85 to 9.97 weight percent of water had a maximum adsorptive capacity for SO_2. The water affected the size of the pores in the gels. He also reported that the adsorption rate for SO_2 decreased markedly if air was present, but did not run tests on air containing varying amounts of water. Ficai (117) claims that gelatinous silica has a lower saturation value for SO_2 than alumina or active carbon, but that it is superior because it is more easily activated by high temperatures or sweeping gases and is cheaper.

The literature on silica contains a great number of references dealing with the manufacture and properties of vanadium on silica gels. These are used primarily to promote the oxidation of SO_2 to SO_3 for the manufacture of sulfuric acid. The pertinence of these references to this study is questionable. These materials are inactive below 400°C and show maximum activity at between 450 and 500°C. This is believed to be the temperature region in which the vanadia is in a molten or liquid form. These references might be pertinent to two-stage operation studies in which the SO_2 in the flue gas would first be oxidized and then the SO_3 would be adsorbed. Although their value in this investigation is doubtful, many of these references were included in the bibliography because of possible leads which they might furnish in allied investigations.

Because of the promise of developing doped silica gel which could adsorb relatively large amounts of SO_2 and other oxides of sulfur, it has been recommended that doped silica gels be included in the studies to be conducted under Phase II.

I. Hydroxides

Several hydroxides have been mentioned for their ability to remove sulfur oxides from gas streams. Perry (277) reported that a ferric hydroxide gel adsorbed an amount of SO_2 from an SO_2-air mixture equal to 10.9% of its weight. Japanese patent 11648/68 (521) mentions ferric hydroxide as being a satisfactory adsorbent for SO_2 while Belgium patent 692,466 (567) states that iron hydroxide is only one of a number of hydroxides which are suitable for adsorbing SO_2. This patent claims that hydroxides of aluminum, manganese, alkali metals and alkali earth metals are also adsorbents for SO_2. Japanese patent 14712/67 (524) also mentions the use of manganese hydroxides as an adsorbent for SO_2.

A Russian paper claims that magnesium hydroxide is a reasonably good adsorbent for SO_2 and that its activity can be increased markedly by hydrothermal treatment at pressures of from 5 to 20 atmospheres (288). According to this abstract, the hydrothermal treatment increases the specific surface and porosity of the magnesium hydroxide and causes a decrease in the effective pore radius of the sample.

All the references described above use the hydroxide in the dry state. However, there are many references which use aqueous suspensions of insoluble metal hydroxides to adsorb sulfur oxides. Hydroxides mentioned in patents as being SO_2 adsorbents include magnesium hydroxide, calcium hydroxide, zinc hydroxide, and iron hydroxide. These data are reported in British patent 708,095 (564) and 680,868 (502); and in Czechoslovakian patent 90,701 (500). The use of an aqueous suspension of magnesium hydroxide to adsorb SO_2 from sintering gases is claimed in a Russian paper (281).

Because of ferric hydroxide's reported ability to remove approximately 11 weight percent of sulfur dioxide from an SO_2-air mixture, work on Phase II should evaluate its use as a potential adsorbent for SO_2. Ferric hydroxide is not a stable material and is not an article of commerce. Therefore, considerable attention will have to be given to its mode of preparation. The preparation used by Perry (277) will be examined, as will the types of ferric hydroxide referred to in other references. A recent Russian patent (218,130), describes the preparation of an active ferric hydroxide base catalyst for the low temperature conversion of ortho-hydrogen to para-hydrogen (505). This process involves the precipitation of ferric iron with sodium hydroxide at approximately room temperature, after which the ferric hydroxide is activated by being kept in a vacuum at 100 to 150°C. This procedure should be tested on some samples of ferric hydroxide used in SO_2 adsorption studies.

The use of an alkaline suspension of ferric hydroxide for SO_2 adsorption is claimed in two patents (512, 492). The use of an alkaline suspension would be particularly beneficial if the adsorption mechanism consisted of dissolving the SO_2 in the aqueous medium and then having the dissolved SO_2 react with the insoluble solids. The use of an alkaline suspension of a hydroxide might be especially attractive if the alkalinity could be imparted to the solution by the fly ash which would be removed by the scrubbing solution. Many fly ashes are known to be basic and to contain relatively large amounts of lime. These might yield a solution sufficiently alkaline to aid the scrubbing of SO_2 with insoluble hydroxides.

SECTION III - REVIEW OF SOLID-GAS CONTACTORS

A thorough review of the literature reveals five processes within the scope of this contract that are in either the pilot or demonstration plant stage of development. In Appendix I detailed process studies of each of these are developed. Presented are flow sheets, material and energy balances, state of development, economics, chemistry, and problem areas to the extent that this information is available. In certain cases, some information was inferred from limited data, and engineering judgement.

The five processes under development are:

- Reinluft
- Sulfacid
- Hitachi
- Grillo
- Still

Of these, the first three employ carbon as an oxidative sorbent. The latter two employ metal salts as sorbents. Schematic flow sheets for each of these processes are shown in Figures 3 through 7 in Appendix II.

A. Carbon Based Processes

Activated carbon is one of the materials widely used as a sorbent for polar gases. The effectiveness of active carbon as a general purpose sorbent correlates well with surface area. There is virtually no correlation, however, between surface area and the effectiveness of various carbons as SO_2 sorbents from stack gases.

Furthermore, the sorptive capacity of carbon for SO_2 from stack gas is several fold greater than its capacity to sorb pure SO_2 at the same total pressure. Juntgen, (175 thru 180) among others, has performed definitive studies which show that the high capacity of carbon for SO_2 from stack gas is the result of a complex series of chemical reactions which consume water and oxygen as well as sulfur dioxide.

Sulfur dioxide is adsorbed on the surface of carbon from the stack gas. Even at saturation, only about 0.5 to 1 percent by weight can be adsorbed by a carbon of surface area approaching 1000 square meters per gram. The carbon surface also adsorbs other gases from the stack gas. Water, carbon dioxide and oxygen are all adsorbed in competition with SO_2.

Certain sites on the carbon surface hold localized unpaired electrons. These sites, which can be detected by electron spin resonance Spectroscopy, are active catalysts for the combination of the sorbed sulfur dioxide with sorbed oxygen. This reaction produces sulfur trioxide as a sorbed species. This, in turn, reacts with sorbed (or gaseous) water to form sulfuric acid.

Even at usual stack gas temperatures, the sulfuric acid is below its dew point. Accordingly, it forms a condensed phase within the pore structure of the carbon. The acid is held within the pore structure of the carbon by capillary forces.

The acid too establishes an equilibrium with the water vapor in the stack gas. Typically, the acid sorbed within the pore structure contains about 30 percent water by weight (1.8 moles H_2O per mole H_2SO_4).

In summary, carbon is functioning as an oxidation catalyst and as an inert matrix to hold the sulfuric acid that is formed on oxidation. Since it is not behaving as an adsorbent, this sorption process cannot be viewed in terms of the conventional concepts of adsorption which are usually applied to sorption of gases on carbon and the concept of an equilibrium sorption isotherm for SO_2 from stack gas on carbon is meaningless. Instead, the process must be approached in terms of the concepts of chemical kinetics in which saturation of the carbon is the filling-up of the carbon with sorbed acid. The active surface of the carbon occludes acid and so decreases the rate of sorption.

The kinetics of the overall reaction (sorption) are interesting and important in themselves since there is a change in the rate controlling step in the temperature range of interest. At low temperatures, the surface is essentially saturated with SO_2 and oxygen, and the rate is limited by the kinetics of the SO_2 oxidation step. As the temperature is increased, the overall reaction rate increases.

As the temperature is increased further, this rate levels out; and eventually begins to fall. This occurs because the sorption of SO_2 on the carbon surface decreases strongly with increasing temperature. Eventually, a point is reached at which the sorbed SO_2 concentration on the surface of the carbon is so low that it limits the rate of the overall reaction which achieves a maximum rate at temperatures between 200 and 250°F.

The regeneration of saturated carbon is also a complex process. To date, two methods have been used. The simplest of these is washing. Water is trickled through a bed of saturated carbon. The water leaches the acid from the carbon particles. Unfortunately, the maximum acid concentration achievable in the leach liquor is rather low. The hydraulics of the flow through the leaching bed seem to be the source of the problem. At an acid concentration between ten and twenty percent, the liquor no longer wets the carbon particles, and satisfactory leaching rates are not attainable. Both the Hitachi and Sulfacid processes rely on leaching for regeneration.

The second regeneration method is thermal regeneration. The sulfuric acid saturated carbon is heated to a temperature at which the carbon reduces the sulfuric acid forming sulfur dioxide, carbon dioxide, and water. Chemically, the process appears fairly complex. Again, Juntgen and co-workers have performed the most definitive work.

The regeneration process does not proceed at a reasonable rate below about 650°F. In the course of heating the saturated carbon to this temperature, the sorbed acid is concentrated to essentially pure H_2SO_4.

It is interesting to note that the onset of the regeneration process corresponds to the onset of the dissociation of sulfuric acid into water and sulfur trioxide. (See Appendix II, Table XIX.) It appears then, that the first step in regeneration is

$$H_2SO_4 \rightarrow SO_3 + H_2O$$

At a temperature of over 600°F, one suspects that the carbon surface retains very little of the sulfur trioxide.

The second step appears to be the reaction of gaseous sulfur trioxide with the carbon surface to form sulfur dioxide and the carbon "surface oxide". This appears to be a covalently bonded oxygen atom attached to a carbon atom on the surface which is still bonded to the carbon matrix by a pair of covalent bonds.

The surface oxide site apparently can react further with a second sulfur trioxide to form carbon dioxide and sulfur dioxide.

$$SO_3 + C \rightarrow ({>}C{=}O) + SO_2$$

$$({>}C{=}O) + SO_3 \rightarrow CO_2 + SO_2$$

There are several observations that support this mechanism. Under certain conditions, it is reported that the majority of the sulfur values can be recovered as sulfur trioxide. This suggests that the trioxide is the initial desorbed product.

The other observation is that virtually no free carbon monoxide is produced on regeneration unless the carbon is heated to temperatures well above 900°F.

Such a high temperature treatment is recommended as a post treatment for the sorbent after regeneration. This is claimed to enhance the sorption activity of the char on the next sorption cycle. There is ample evidence that excessive coverage of the carbon surface by surface oxide makes it inactive in catalyzing the oxidation of sulfur dioxide during sorption.

Active carbons with low ash contents lose their activity for SO_2 sorption on recycling, either with leaching, or thermal regeneration. This is attributed to surface oxide formation. It can be avoided by adding transition metals or iodates to the carbon. Similar problems are not always encountered with carbons having high ash contents, perhaps because of their natural metal content.

1. Reinluft Process[1]

The Reinluft process uses moving beds of lump char for both sorption and regeneration. To hold pressure drop to a minimum, the particles are one to two centimeters in diameter. Because the particles are large, there is a major mass transfer limitation on the rate of sorption. Solids residence times of several hours are required to saturate the char. Ultimately, this means that a very large volume of equipment is required. To maintain control over the solids flow, a high multiplicity of units is needed.

To date, the most severe problem with the Reinluft process has been autoignition of the char beds. No problem has been encountered on startup, but after a week or so of operation, the beds have ignited. This appears to be the result of activation and consumption of the char during regeneration. As activation proceeds, the surface area (and perhaps the specific activity of the char) increases. The consumption of carbon decreases the physical strength of the char, and its thermal conductivity. A point is reached where the heat of chemisorption. -- both of SO_2 and also of oxygen -- cannot be effectively dissipated and eventually the temperature inside the char lumps exceed the autoignition temperature of the activated char. Efforts to correct this condition by reducing the temperature of the char recycled from the regenerator were unsuccessful. The point of ignition appears to have been in the area where the regenerated char was injected.

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[1] Flow diagrams and further descriptions of each process are presented in Appendix II.

The most recent Reinluft Pilot at Leunen appears to have operated without ignition problems. The oxygen level in the stack gas there -- three percent -- is significantly lower than that in some of the earlier unsuccessful pilot plants. The effectiveness of the Leunen installation in removing SO_2 was somewhat below design.

It has been reported that Reinluft G.m.b.H. has abandoned this process and has sold all rights to Bergwerke Verbrand, a company with close ties to Bergbau-Forschung G.m.b.H. - Essen, the German coal research institute. This is based on a private communication and has not yet appeared in the literature.

2. Sulfacid Process

The Sulfacid process is under development by Metallgesellschaft A.G. - Frankfurt (Lurgi). This process is also sometimes referred to in the literature as the "Stratmann process". This is the most advanced of the processes, in that there are commercial installations in service with several years' operating experience.

In the strictest sense, the Sulfacid process is not a sorption process. The sorption and regeneration by leaching occur simultaneously. Hence, the carbon "sorbent" is really functioning only as a catalyst bed. The stack gas is first saturated with water. Then it flows downward through a fixed bed of active carbon one or two feet thick made up of carbon particles about 5-6 millimeters in diameter. Water is also sprayed over the top of the bed and is allowed to trickle through the bed. The resulting acid is no more concentrated than twenty percent by weight.

Lurgi proposes that this acid be contacted with incoming hot gases to cool and partially saturate the gas while concentrating the acid. This process modification has yet to be demonstrated in a commercial installation.

The Sulfacid process is not particularly well suited to large power plants. The water requirements are very large. About sixty pounds of water are used per pound of sulfur dioxide recovered. Ninety-five percent of the water used is evaporated into the stack gas. The stack plume is not only moist, but also fairly cool. The optimal operating temperature of the absorber bed appears to be about 140°F. The total volume of active carbon bed required to treat the total power plant gas is quite large.

While the process does not lend itself to power plant applications, it is ideally suited to sulfuric acid plant waste gases. There the dilute acid product is no problem, since it can be fed as part of the makeup to the primary scrubber and the exhaust gases generally are hot enough for adiabatic saturation with water. Hence, the only operating problem is the fairly high water requirement.

3. Hitachi Process

The Hitachi process uses a number of packed carbon beds as chromatographic absorbers. When the saturation front is about to break through one bed, it is switched to a washing phase. When the bed is fully leached, it is dried by passing a fraction (25%) of the incoming flue gas through it to dry out the carbon in preparation for a second absorption cycle. A two megawatt pilot plant was satisfactorily operated at Tokyo Electric's Gai Plant and a larger demonstration plant is to be constructed. The operating cycle is 60 hours long with 30 hours of adsorption, 10 hours of washing and 20 hours of drying. In the preferred method of practice, the washing operation is carried out countercurrently. A nearly exhausted tower is washed with incoming water while a tower just off of the sorption cycle is washed with leach liquor from other towers. Acid concentration is still limited to 20 percent, however.

Because packed beds are used, the carbon particles must be fairly large (1/2 to 1 inch) to hold the pressure drop down. Diffusion into the carbon particles then limits the rate of adsorption. Large inventories of carbon are required, and therefore bulky equipment must be used. The chromatographic mode of operation further dictates that a large fraction of the carbon is performing no useful function at any given instant. The towers must be constructed of acid resistant materials, and extensive duct and damper systems are required to route the gas to the various towers.

B. Non-Carbon Processes

1. Grillo Process

The Grillo process is being developed by A.G. fur Zinkindustrie vorm Wilhelm Grillo. Their interest is primarily in the treatment of waste gases from their smelters and acid plants. In this context, the process makes good sense since the equipment is normally a part of the smelter operation. For power plant applications, the complexity of the operation and the number of process steps involved would seem to rule this process out.

The sorbent is a mixture of an alkaline earth hydroxide and a transition metal (higher oxidation state) oxide. The preferred sorbent is magnesium hydroxide and manganese dioxide. Sorption occurs in a spray drier into which a slurry of the absorbent is sprayed. Two spray driers in series are used on the gas side. The sorbent is recycled extensively within each drier (dry sorbent is reslurried and returned to process). A small portion of the sorbent is moved countercurrent to the gas each cycle. The spent sorbent, containing about 40 percent sulfur dioxide by weight, is regenerated by a reductive roasting followed by quenching, clarification to remove dust and ash, filtration, and drying.

The purified flue gas leaves the process saturated with water at 120°F. Reheat of this gas would almost certainly be required. The water consumption would be at least 50 pounds per pound of sulfur dioxide removed. This estimate includes only losses to the waste gas. Even assuming that water is readily available, there would be a cost of at least 20¢ per ton of coal burned for power just to provide the process water.

To summarize, the Grillo process simply does not seem to lend itself well to power plant applications. The problems are in both sorption and regeneration, but probably the greatest problem is regeneration which is a chemically complex series of a number of process steps. While the utility industry may reluctantly accept an automated two step process as a necessary evil, it is questionable that they would consider a six step process.

In criticizing the process, it is important not to lose sight of its two desirable aspects. One is the high capacity sorbent (40% by weight SO_2) and the other is the use of dispersed solid contactors.

2. Still Process

Germany's Firma Karl Still is developing a process which uses the fly ashes from the combustion of certain lignites as sorbents. The ashes which prove active are rich in calcium and sodium and they also contain significant quantities of transition metal oxides. Only the ashes which have been hydrated to produce alkalai and alkaline earth hydroxides are active.

In the Still pilot plant, the sorbent contacts the stack gases in a series of three transport contactors. The solids are mixed with the gas, conveyed with it through an inverted "U", and are collected in a cyclone. Most of the solids are recycled to the same contactor, but some are advanced to preceeding contactors.

The solids are regenerated thermally. Regeneration is only partial, however, Some of the sorbent is bound as sulfate which is not regenerable. Thus, after a few cycles, the sorbent is spent and must be discarded.

Unfortunately, coals which yield active ashes do not appear to be widespread. This, of course, raises the question whether inactive ashes can be made active. As a first attempt, Still is supposed to have tried to use hydrated lime to start up it's pilot plant while awaiting deliveries of ash. These preliminary results are reported to have been discouraging. Further runs with hydrated limes are planned, however.

The specific sorbent employed by the Still process is not a particularly good one. It is poorly distributed geographically, and it is rapidly consumed by the process. The engineering, however, appears to be excellent. The multiple-stage transport contactor should be a promising device in conjunction with other sorbents.

C. Comparison of Existing Processes

The five existing processes are compared in Table X on a number of points. The first three items of comparison provide a capsule process description, the others the bases of comparison.

1. Costs

In this compilation, capital costs for large base load power plants (approximately 800 megawatt), are presented in $ per kw of installed capacity. The operating cost estimates are in mills per kwh. By-product credits are not included in the operating costs, and by-product processing equipment is not included in the capital. Vendors estimates are presented in parenthesis; those not in parenthesis are from Katell (188, 189). It is interesting to note for the Reinluft process the wide disparity between the cost figures provided by Reinluft G.m.b.H. and those estimated by Katell. Katell's figures are nearly 50 percent higher for the capital and almost 100 percent higher for operating cost. The greatest point of difference is sorbent cost. Johswich estimates the sorbent at $20 per ton while Katell estimates it at $80 per ton. The latter figure seems far more reasonable. In a second independent estimate, Zentgraff (447) estimates capital and operating costs still higher than Katell's.

In general, the vendors cost estimates must be regarded as absolute minimum values. However, Lurgi figures should be fairly accurate since they are based on operating experience. The figures for the Still process must be regarded as highly tentative since they do not reflect pilot-plant experience.

TABLE X

COMPARISON OF PRESENT INORGANIC SOLID SORPTION PROCESSES

Process	Reinluft	Sulfacid	Hitachi	Grillo	Still
Sorbent	Semicoke	Active Carbon	Active Carbon	$Mg(OH)_2 + MnO_2$	Lignite Ash
Sorption mode and sorbed species	Oxidative sorption H_2SO_4	Oxidative sorption N.A.	Oxidative sorption H_2SO_4	Reaction $MgSO_3 + MnSO_4$	Reaction $CaSO_3/CaSO_4$
Regeneration Mode and Byproduct	Thermal SO_2	Washing 60-70% H_2SO_4	Washing 10-20% H_2SO_4	Thermal, Reductive SO_2 or S, COS, H_2S	Thermal SO_2
Capital Cost, large plant \$/kw (vendors est.)	\$17.80 (12.8)	(\$14.50)	\$10.00	?	(\$5.00)
Operating Cost mills/kwh no B.P. credit (vendors est.)	0.86 (0.44)	(0.4)	0.45	?	(0.5)
Scale Factor	0.9 to 1.0	0.8 to 1.0	0.9 to 1.0	0.8 to 0.9	0.7 to 0.8
Adapt. to Existing Plant	Difficult	Difficult	Difficult	Difficult	Possible if temp. high enough
Stack Gas	215°F 8% H_2O	<150°F Sat.	250°F 10% H_2O	Est. 120°F Sat.	300°F 8% H_2O
Operability	Ignition problem seems under control. Demonstrated recovery only 75%	Operating very well on H_2SO_4 tail gases >99% recovery	Problems with extensive ducting and dampers	Unknown. Different contactors being used in pilot	Source of absorbent and disposal of spent absorbent are problems
Credits for Precipitator	Yes	Optional	No	Yes?	No (possibly negative)
Stack height	Yes	No	Yes	No	Yes

Furthermore, the operating costs will be almost completely dominated by sorbent costs which will vary greatly from point to point. The vendor's estimate is presumed to be based on a power plant burning the lignite supplying the active ash.

2. Scale Factor

The scale factor serves simply to emphasize that the fixed bed contactors used in all three carbon processes will require a very high multiplicity of units. Hence, there will be no particular cost advantage for large installations. This should not be so large a problem with dispersed phase absorbers, such as those used in the Grillo and Still processes. This is reflected in the lower scale factor applied to these two processes.

3. Adaptability to Existing Plants

Of the processes considered, only the Still process offers a reasonable possibility of installation in an existing plant. The bed area for the three carbon based processes is prohibitive, as would be the ducting. The Grillo absorbers might be accommodated, but the regeneration system would have to be located off site. With the high sorption capacity of the Grillo process, off-site regeneration might be feasible.

4. Product Gas

The gases generated by both the Sulfacid and Grillo processes are cool and saturated with water, and both would require reheat before they could be discharged. Reheat costs have not been added into the operating costs. The product gases from the other three processes, the Hitachi, Reinluft and Still processes, could be discharged to the atmosphere without problem.

5. Operability

This section highlights the operating problems of the processes. The Reinluft process has been plagued by ignition of the sorbent bed and by low sorption efficiencies. The literature does not report any major problems encountered in operating the Lurgi process. Sulfur dioxide removals have been in excess of 99 percent. The only major operating problems of the Hitachi process have been connected with the maze of ducts and dampers required for its practice. Of the Grillo process, the use of an entirely different contactor in the pilot than was used in bench scale work was reported but not accounted for. There is sufficient information to show that the spray dryer being used in the pilot could not have been modeled on the bench; so this may not indicate trouble. So far as is known from available information, no operating problems have been experienced by the Still process, but there are certainly logistic problems of supplying fresh sorbent and disposing of spent sorbent.

6. Credits

In evaluating these processes in the context of a new power plant, the question of credits for elimination of the precipitator and for using shorter stacks arises. Credits for elimination of the precipitator are claimed by the Reinluft process. In the Sulfacid process, where a venturi scrubber is used to contact the entering gases with acid, the precipitator can be eliminated. This is accomplished by use of an acid clean-up system which might be more expensive than the precipitator. The effectiveness of the proposed separation step of removing the ash from the sorbent, however, appears questionable at best. Dust removal as a post treatment in the Grillo process is required in any event. Thus, for this process, the question is simply whether one or two precipitators would be required.

The possibilities of stack height reduction are much more difficult to assess. There are a great many degrees of reduction of stack height. In general, we have indicated that stack reduction is not possible in the case of the Sulfacid and Grillo processes, which produce rather cool plumes. This assumes that reheating is not practical.

It seems certain that any process that takes credit for stack reduction must discharge a buoyant plume, and must reduce emissions, not only of SO_x, but also of NO_x, fly ash, and hydrocarbons to a very low level.

7. Summary

In summary, none of the five existing processes appears to be the ideal answer for pollution control. The Sulfacid process appears to be well suited to treating sulfuric acid tail gases. The Grillo process appears suited to large metallurgical smelter operations, but poorly suited to other emission sources. The Still process appears to be a good one where active ashes are available.

D. New Developments

A survey of the current literature reveals a number of clues to work in progress. It seems evident to many observers that the fixed bed absorber is poorly suited to treating stack gas. Sorbent particles must be large to keep pressure drop down. Large sorbent particles react slowly because diffusion distances inside the particle are long.

The trend now is toward dispersed solid contactors. These may be of three types:

- multiple thin fluid beds
- highly recycled conveyed solids systems
- highly recycled raining beds

Each of these has its advantages.

The fluid beds have the advantage of being countercurrent contactors. The gas velocities are fairly low (1-5 fps) so contacts are very brief. These devices also waste a good deal of pressure head across the multiple grid plates.

The raining and conveyed beds both require a fairly large contactor (1-2 seconds of gas hold-up), and both are troubled to some extent by segregation of gas and solids. In both cases, solids residence times, per pass, are low. This means that solids having moderate to high sorptive capacity (10% SO_2 by weight) must be recycled many times through the contactor.

All three of these contactors expose the sorbent to a severe mechanical battering. In such contactors then, the attrition resistence of the sorbent becomes very important. Hitachi has recently patented a raining bed contactor which has interval baffle plates to continuously remix the solids and gas, and to slow down the rate of passage of solids through the contactor. This contactor would be well suited to sorptive carbons. (It is tempting to speculate that Hitachi is working on a process using granular carbon, and a dispersed phase contactor.)

The conveyed contactor used by the Still process is an original design that should find extensive use in processes using sorbents other than active ashes.

Comments in recent articles by Juntgen et al. suggest that new possibilities in the regeneration of carbon sorbents are being explored at Bergbau-Forschung. He suggests that they have had success in defining conditions where sulfuric acid will dissociate to form sulfur trioxide and water, but where most of the sulfur trioxide can be recovered without reduction. Unfortunately, this can probably be done only at low SO_3 partial pressures and, for this reason, it may not be commercially useful.

The other process possibility is that of reducing sorbed sulfuric acid to SO_2 with gaseous reductants rather than with carbon. Juntgen hints that Bergbau is following up on this. The idea appears so promising that we recommend work in the U.S. on this regeneration concept. (See above.)

The possibility of a non-destructive process using a carbon sorbent is appealing, but it may not be possible to find conditions coupling a satisfactory rate of reaction with the selectivity desired.

Carbon is such a popular sorbent because its mode of sorption leads to an inherently high capacity limited only by the porosity of the carbonaceous mass. The disadvantage of this process is the fact that the carbon degrades on regeneration, or produces a relatively low-valued by-product. The preceeding discussion has focused on two possible means of circumventing the degradation problem. A third way is to substitute some other porous material which is inert to sulfuric acid for carbon. Within this matrix material a suitable catalyst for SO_2 oxidation at stack gas temperatures should be deposited. The catalyst material would preferably, but not necessarily, be soluble, in sulfuric acid. Should the material be acid soluble, no loss would be expected since the acid solution would be retained within the porous matrix of the support. For the support matrix, we suggest silica in the form of silica gel.

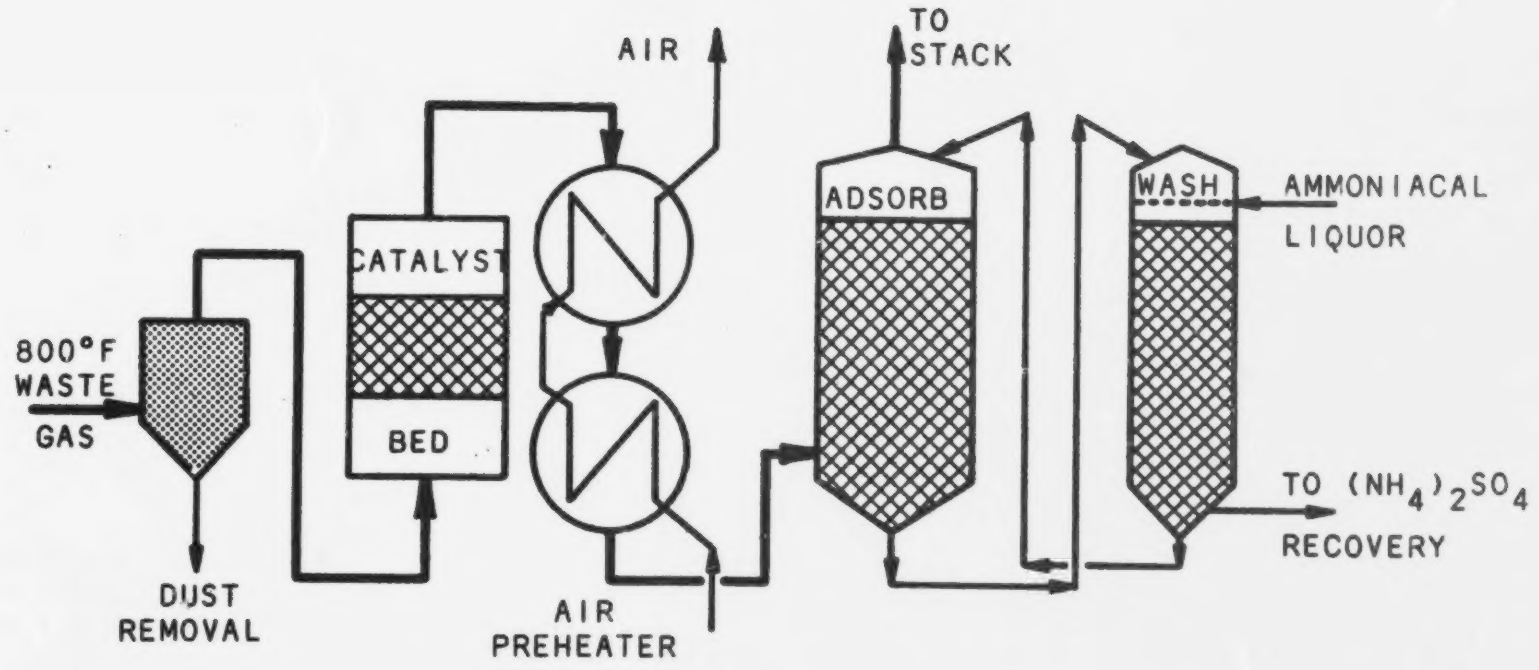
The regeneration of such a sorbent could be either thermal, with the production of SO_3 and water vapor at 700 to 900°F, or reductive, with the production of SO_2. Leaching might also be a possible means of acid recovery, and higher acid concentrations might be possible with a more hydrophylic sorbent. Leaching could not be used if the oxidation catalyst is at all acid soluble. We recommend that a search be made for suitable catalystic materials to make a silica sorption process a reality.

An SO_x removal process in this classification is the subject of a recent patent issued to Juntgen (468) and assigned to Bergwerke-Verband. The process scheme is shown in Figure 2, page 55. Dust is removed from the stack gas in a high temperature precipitator. The gases then pass through a vanadium pentoxide bed at about 800°F. Thereafter, they are cooled in the air preheater of the plant to 300 to 400°F. The gases then pass through a moving bed absorber, where the sulfuric acid mist is sorbed onto coke. The coke may be regenerated by washing or by a thermal-reductive process, as in Reinluft.

This process would seem to offer no particular advantages over either the "Pennelec" (catalytic oxidation) or Reinluft processes, and indeed serves to combine the worst of both. It is included here solely for completeness.

Figure 2

Bergwerke-Verband SO_2 Removal Process


800°F
WASTE
GAS
DUST
REMOVAL
CATALYST
BED
AIR
AIR
PREHEATER
TO
STACK
ADSORB
WASH
AMMONIACAL
LIQUOR
TO $(NH_4)_2SO_4$
RECOVERY

E. Process Economics

The process economics presented in Table X and in Appendix III are strongly influenced by sorbent costs. This is true of both capital and operating costs. As we have mentioned earlier, the greatest disparity between Reinluft's estimate ($12.80 per installed kw, 0.44 mills per kwh) and Katell's estimate for the Reinluft process ($17.80 per kw 0.86 mills per kwh) is in the cost of sorbent. This influences capital through sorbent inventory, as well as operating cost through sorbent consumption. Sorbent costs are important in the Alkalyzed Alumina Process as in virtually all of the inorganic solid-gas processes. In all these, process variables such as sorbent attrition, chemical degradation of sorbent, and sorbent capacity are required to provide more than rough order of magnitude costs.

The literature search on sorption by inorganic solids has shown a considerable need for more base information. Data indicating sorption under anything approximating stack gas conditions have been rare enough. Virtually no data on attrition and sorbent degradation have been encountered on any sorbent other than carbon. Hence, it is not really possible to estimate even hypothetical costs with sufficient accuracy to justify the exercise. We can, however, use process economics to indicate the economically sensitive areas so that preliminary process studies can be concentrated there.

The sorbent cost may vary over several orders of magnitude. Minerals may be mined and crushed to produce crude sorbent for well under $5.00 per ton. Minerals or waste materials, processed through fairly large plants, could run as much as $20 per ton in granular form. At the other end of the cost scale, one can easily propose sorbents that would cost well over $100 per ton.

Processes with expensive sorbents must have very low sorbent losses and very small capacity losses on processing. Processes with very inexpensive sorbents can operate with high losses, but under such circumstances, sorbent disposal may become a severe problem. Such is the case for the limestone scrubbing processes.

Sorbent capacity interacts strongly with the complexity and cost of the regeneration process. A very high capacity sorbent (40-50% by wt. SO_2) can carry a relatively high unit regeneration cost. A low capacity sorbent (<3% SO_2) must have a simple, inexpensive regeneration process. Quite obviously, there is a wide range in between.

The type of contactor used is an important economic variable. In this area, we have performed some economic analysis. As was pointed out earlier, we feel that the dispersed phase contactor is most desirable if attrition losses can be held low enough. To hold attrition to reasonable levels, one approach is to reduce the kinetic energy of the particles. This greatly decreases impact forces. As one goes to smaller and smaller particles, however, mass transfer becomes limiting since the relative velocities between solids and gas also decrease. Solids recovery becomes a more severe problem with decreasing particle size also. In general, sorbent particles in the 1 to 0.1 mm range represent a reasonable compromise. Such particles have terminal velocities in the range of 3 to 10 feet per second, and can be readily recovered in centrifugal separators. Fragments from these particles are large enough to be recovered in cyclones.

If it is assumed that the sorption is diffusion controlled, then in a five second contact time, with particles of the above size range, over ninety percent of the sulfur dioxide can be removed. This contact time can be achieved in a 125 foot high inverted "U" column, or in a 250 feet high raining bed column with a 40 foot per second gas velocity, and a solids loading of one pound per 40 cu. ft. of reactor. This is a reasonable solids loading for such a system. The pressure would be under 5" of water for the raining bed system, and less for the conveyed solids system.

While the gas could be desulfurized to a satisfactory level in this system, the sorbent would pick up only about 0.005 pounds of SO_2 per pound per pass. Thus, about 25 passes would be necessary to saturate the sorbent if it were carbon.

For an 800 mw power plant, a stack about 35 feet in diameter would be adequate. For such a system, we estimate the following costs:

		Installed Costs
Contactor		$ 360M
Cyclone Separators		$ 300M
Surge Bins		$ 60M
Draft Fan Drive 5" H_2O		$ 35M
Installed Equipment		$ 755M
Piping & Ducting	40%	
Instruments	20%	
Engineering	30%	
	90%	$ 685M
Total Cost		$1440M
Contingency	15%	$ 212M
Total Cost		$1652M

The erected contactor costs are based on analogy with steel stack costs (1,2). The remaining costs were based on Dryden and Winfield (3) and Vilbrandt and Dryden (4).

This, we feel, is a reasonable estimate for the capital costs of the adsorber including the draft fan. The power consumption by the driver for the booster fan is estimated at about 500 kw of power (Chilton).

(1) Stankiewicz, E.J., "How To Estimate Stock Costs" Chemical Engineering, June 1955

(2) Chilton, C.H., "Cost Engineering in the Process Industries" McGraw-Hill Book Co., Inc. 1960

(3) Dryden, C.E., and Winfield, M.D., "Chemical Engineering Costs" Special Report 29 - Engineering Experiment Station, Ohio State University, Columbus, Ohio 1963

(4) Dryden, C.E., and Vilbrandt, F.C., "Chemical Engineering Plant Design", 4th Ed. McGraw-Hill Book Co., Inc. 1959

SECTION IV - THERMODYNAMICS OF SULFUR DIOXIDE SORPTION

The thermochemistry of a number of potential SO_2 sorption reactions was calculated for a wide variety of common elements. A list of potential SO_2 sorption reactions was formulated. Thermochemical data, specifically

- Enthalpy of Formation at 298.15°K from the elements,
- Absolute Entropy of Formation at 298.15°C.,
- Heat capacity equations, or alternatively, heat capacity data to which an equation could be fitted,

were then sought. In so far as possible, data were taken from the Tracor, Incorporated, under Contract PH-86-68-68. Table XI shows the data that were collected on metal compounds and the source of each item of data. The data themselves are tabulated in Appendix II. In addition to the data on metal compounds, data were assembled on a number of non-metal compounds that were involved in the postulated chemical reactions. These data too are listed in Appendix II.

Where heat capacity equations were fitted to heat capacity data, a four constant form of equation was used. This form of equation was chosen because computer studies indicated that the fit was as good as or better than that attained with three constant equations.

In general, the four constant equation was superior for the case of gases containing four or more atoms. For simpler gases and solids, the fourth constant had no effect on the correlation The C_p equation used throughout was

$$C_p(T) = a + bT + cT^2 + d/T^2$$

By integration of this equation, closed expressions for enthalpy and entrophy were obtained.

$$H_f(T) = H_f^\circ + a(T-T^\circ) + \frac{b}{2}(T^2-T^{\circ 2}) + \frac{c}{3}(T^3-T^{\circ 3})$$
$$-d(1/T - 1/T^\circ)^2$$

$$S(T) = S^\circ + a \ln(T/T^\circ) + b(T-T_o) + \frac{c}{2}(T^2-T_o^2)$$
$$- \frac{d}{2}(1/T^2-1/T^{\circ 2})$$

TABLE XI

SOURCES OF THERMODYNAMIC DATA FOR METAL SALTS

	MET			$O^=$			OH^-			$S^=$			$S_2^=$			$SO_3^=$			$SO_4^=$			$CO_3^=$			$SiO_3^=$			$PO_4^=$		
	H	S	Cp	H	S	Cp	H	S	Cp	H	S	Cp	H	S	Cp	H	S	Cp	H	S	Cp	H	S	Cp	H	S	Cp	H	S	Cp
Li^+	T	T	T	T	T	T	L	L	L							T	T	T	T	T	T	T	T	T	L	L				
Na^+	T	T	T	T	T	T	L	L	L	L	L	K	L			T	T	T	T	T	T	T	T	T	L	L	L	L		L
K^+	T	T	T	T	T	T	N	L		N	L		N			T	T	T	T	T	T	T	T	T						
Mg^+	T	T	T	T	T	T	L	L	L	T	T	T				T	T	T	T	T	T	T	T	T	L	L	L	L		L
Ca^+	T	T	T	T	T	T	L	L	L	L	L	L				T	T	T	T	T	T	T	T	T	L	L	L	L	L	L
Ba^+	T	T	T	T	T	T	L		K	N						T	T	T	T	T	T	T	T	T	N			N		
Cr+3	T	T	T	T	T	T	N																							
Mo+4	T	T	T	L	L	L				L	L	L																		
+6				T	T	T				L	L																			
Mn+2	T	T	T	T	T	T	L	L		T	T	T				T	T	T	T	T	T	T	T	T	L	L	L	N		
Mn+3				T	T	T	N												T	T	T									
Fe+2	T	T	T	T	T	T	L	L		T	T	T	T	T	T				T	T	T	T	T	T	L	L	K	N		
+3				T	T	T				N						T	T	T	T	T	T	T	T	T						
Co+2	T	T	T	T	T	T	N			N	M	N							T	T	T	T	T	T						
Co+3							N			N																				
Ni+2	T	T	T	T	T	T	L	L		L	L	L							T	T	T	T	T	T						
Cu+1	T	T	T	T	T	T				T	T	T				T	T	T	T	T	T	T	T	T						
+2				T	T	T	N			L	L	K				T	T	T	T	T	T	T	T	T						
Zn+2	T	T	T	T	T	T	L	L	N	T	T	T				T	T	T	T	T	T	T	T	T	L	L				
Sn+2	T	T	T	T	T	T	B	B		T	T	T																		
+4				B	B	K	B			T	T	T				T	T	T	T	T	T	T	T	T						

Key:

T = tracor Data Base
L = Landolt-Bornstein Tabellen
N = NBS Circular 500

B = NBS TN270-2
K = U.S. Bu. Mines Circ. 584
M = U.S. Bu. Mines Circ. 406

Phase transitions were handled, where necessary, by treating the new phase as a separate compound. In general, because the program was essentially a screening program, it was decided not to be concerned excessively with solid-solid phase transitions except where the reaction in question was on the edge of thermodynamic feasibility.

From the enthalpy and entrophy data, free energies of formation were calculated. These were summed to calculate the free energies of reaction. From the free energy of reaction the equilibrium constant was calculated.

$$F_f(T) = H(T) - T\,S(T)$$

$$\Delta F_r(T) = \Sigma_i \nu_i F_{i_f}(T)$$

$$-\frac{\Delta F_r(T)}{RT} = \ln K_a$$

These computations were all carried out with the aid of a digital computer. The programs used are labeled Computer Programs III and IV in Appendix III at the end of this report.

1. Results of Thermochemical Screening

The tables resulting from the thermochemical calculations are presented in Appendix III, Tables XVIII through XLVI. The significance of the computations of course varies from computation to computation. The eight standard reactions that were investigated for all cases for which the data were available together with the expression for the K_p are shown in Table XII.

TABLE XII

Standard Metal Compound Reaction Classes

Class No.	Reaction	Kp
I	$M\ CO_3 + SO_2 \rightleftarrows M\ SO_3 + CO_2$	P_{CO_2}/P_{SO_2}
II	$M\ (OH)_2 + SO_2 \rightleftarrows M\ SO_3 + H_2O$	P_{H_2O}/P_{SO_2}
III	$M\ SiO_3 + SO_2 \rightleftarrows M\ SO_3 + SiO_2$	$1/P_{SO_2}$
IV	$M\ CO_3 + 1/2\ O_2 + SO_2 \rightleftarrows M\ SO_4 + CO_2$	$P_{CO_2}/P_{SO_2} \cdot \sqrt{P}_{O_2}$
V	$M\ (OH)_2 + 1/2\ O_2 + SO_2 \rightleftarrows M\ SO_4 + H_2O$	$P_{H_2O}/P_{SO_2} \cdot \sqrt{P}_{O_2}$
VI	$M\ SiO_3 + 1/2\ O_2 + SO_2 \rightleftarrows M\ SO_4 + SiO_2$	$1/P_{SO_2} \cdot \sqrt{P}_{O_2}$
VII	$M\ S + 2SO_2 \rightleftarrows M\ SO_4 + S_2$	$P_{S_2}/P_{(SO_2)^2}$
VIII	$2MS + SO_2 \rightleftarrows 2MO + \frac{3}{2}S_2.$	$P_{S_2}^{1.5}/P_{SO_2}$

Reaction classes I through III are simple acid replacement reactions; classes IV through VI are oxidative acid replacements. Reaction classes VII and VIII redox reactions in which the oxidation state of sulfur changes. Class VIII is the "modified" Claus reaction.

Since the expression of Kp is different for each reaction, the value of K required for the reaction to be thermodynamically feasible also varies. The values of Kp(minimum) which must be exceeded for reactions of each class to be feasible in a typical power plant stack gas to produce an effluent SO_2 concentration of 300 ppm are shown in Table XIII.

TABLE XIII

Minimum Values of Kp for Thermodynamic Feasibility Accepting SO_2 From Power Plant Stack Gas

Effluent Stack Gas Composition:

SO_2	0.3 mole %
H_2O	7 mole %
CO_2	14 mole %
O_2	3 mole %
N_2	Balance

Reaction Class	Kp Formulation	Kp (Min.)
I	P_{co_2}/P_{so_2}	470
II	P_{H_2O}/P_{so_2}	235
III	$1/P_{so_2}$	3333
IV	$P_{co_2}/P_{so_2}\cdot\sqrt{O_2}$	2700
V	$P_{H_2O}/P_{so_2}\cdot\sqrt{O_2}$	1350
VI	$1/P_{so_2}\cdot\sqrt{O_2}$	19200

The criteria cited are, of course, specific to the effluent gas in question. If the concentration of any of the species is changed appreciably, the Kp (min.) for some, or all of the reactions changes accordingly.

Those reaction classes in which sulfur is a product species (VII and VIII) are not handled so simply. For convenience, the thermodynamics of these reactions have been calculated in terms of the molecular species S_2. Unfortunately, S_2 is not the dominant sulfur species except at the top of the temperature range of interest, i.e., near 1000°K.

The standard state of sulfur in the range of interest is defined as pure condensed sulfur under its normal vapor pressure. From 298 to 393°K, this is solid rhombic sulfur composed of S_8 rings. At 393°K, the solid melts. As the temperature of the melt increases, there is some cleavage of S_8 rings with the subsequent formation of polymers. At still higher temperatures, the polymers degrade to short chains. These occur in equilibrium with S_8 and S_6 rings.

The vapor in equilibrium with the sulfur melt is S_8 at low temperatures. At higher temperatures mixtures of S_8 and S_6 predominate. At temperatures approaching 1000°k, S_2 becomes the dominant species.

If S_2 were a stable gaseous species, then the correct values of K_{VII} (min.) and K_{VIII} (min.) would be 3333 and 0.547 respectively. Since S_2 is a hypothetical state, we must add to the free energy of reaction, the free energy of forming the stable form of sulfur from S_2. The corrected minimum values of K_{VII} and K_{VIII} are presented in Table XIV: These are computed as K_{VII} (min.)* K_{S_2}. Since the numbers cover several orders of magnitude, they are reported as logarithms (base ten).

TABLE XIV

Minimum $\text{Log}_{10}K_p$'s for Reaction Classes VII and VIII

T°K	$\log_{10}K_p$VII	$\log_{10}K_p$VIII
500	-0.6707	-8.0516
600	+1.3666	-4.9957
700	+2.7798	-2.8759
800	+3.8124	-1.3270
900	+4.5952	-0.1528
1000	+5.2054	+0.7625

The ranges of thermodynamic feasibility for the reactions studied are shown in Table XV. For each metal ion, listed in that table, there is a box which contains a + for the reaction if it is favorable from 298.15 to 1000°K, a minus if the reaction is unfavorable over that same range, or a specific temperature range in which the reaction is thermodynamically favored (e.g.<975). It must be appreciated that the ranges indicated are only approximate since the data for many of the compounds are of relatively low accuracy. This is particularly true of the hydroxides and sulfides. A significant number of the fundamental thermodynamic parameters are estimated rather than measured values.

For this reason, we did not build into the thermodynamic computational program the sophistication to accommodate phase transitions. The data presented in Table XV are generally based on room temperature phases. Including the solid-solid transitions would introduce some relatively minor changes in the table.

It must also be realized that a table of this sort is specific to one effluent gas. Changes in the composition with respect to sulfur dioxide, water, carbon dioxide or oxygen would alter the table. Similarly, a change in the total pressure of the systems would result in alteration of the table. For this reason, the tabulation of Kp values in Appendix II should be regarded as the primary source of evaluation data. A check should also be made of the data files in Appendix II to determine if there is a solid-solid phase change in the temperature range of interest.

It is interesting to note that of 81 reactions on which computations were made, only 14 were catagorically unfavorable over the range of interest. This is not to say that all of the other 67 reactions are good candidates for SO_2 sorption. In some, the reactants or products are not themselves stable at the given temperatures and gas mixture compositions. For example, each of the carbonates and hydroxides is in equilibrium with the gas. For each of them, there is a temperature above which they will dissociate to the oxide and either CO_2 or water. The same can be said for product species such as sulfite and sulfate.

Certain of the reactants are hygroscopic. Sodium hydroxide would sorb water strongly from the stack gas at temperatures near the boiling point of water.

The two sulfide reactions, Class VII and VIII, quite obviously yield non-equilibrium states, if there is any oxygen in the gas phase. Oxidation of the S_8 and S_6 ring is a very slow reaction, however. Hence, at temperatures below say 700°K, it may be possible to carry out these reactions without oxidizing the product sulfur to form more SO_2.

TABLE XV

Range of Thermodynamic Feasibility of Various Reactions From 298°K - 1000°K for Typical Stack Gas Composition

Class								
Metal								
Li	<900°K	+	0	+	+	0	0	0
Na	+	+	<650°K	+	+	<950°K	<850°K	-
K	+	0	0	+	0	0	0	0
Mg	-	<375°K	<450	+	+	<850	<750	+
Ca	<575	+	<600	+	+	<950	<750	0
Ba	+	+	0	+	+	0	0	0
Mn+2	<425	0	<575	+	0	<950	<550	-
Mn+3	<550	0	0	+	0	0	0	0
Fe+2	<375	0	<425	+	0	<875	<425	-
Fe+3	<475	0	0	+	0	0	0	0
Co+2	<775	0	0	+	0	0	-	+
Ni+2	0	0	0	0	0	0	<500	-
Cu+2	<575	0	0	+	0	0	-	-
Cu+1	<475	0	0	<750°K	0	0	-	-
Zn	+	0	0	+	0	0	-	-
Sn+2	0	0	0	0	0	0	0	-
Sn+4	<375	0	0	+	0	0	-	<825°K

Key - + = Favorable at all Temp.
0 = No data.
- = Unfavorable at all Temps.
Other entries indicate ranges of favorable values

The tables of Appendix II are but the first step in the thermodynamic characterization of any single reaction system.

Once a specific reactant system has been selected for study, it should be subject to further thermodynamic analysis of the stability of the several reactants and products. The thermodynamics of possible side reactions should likewise be examined if the necessary fundamental data are available.

OTHER REACTIONS

Where the metal atoms involved in reactions have multiple valence states additional reactions become possible. These are reactions in which the oxidation state of both sulfur and the metal are changed.

Of the systems studied, sufficient data to explore the feasibility of such reactions was found for only the compounds of manganese, iron, and copper.

Of the seven reactions of this type studied, all were unconditionally favorable over the entire temperature range (for the stack gas composition used in Table XIII).

The reactions of the iron system give a suitable example. The three reactions studied were:

$$Fe_2(CO_3)_3 + 2SO_2 \rightleftarrows FeSO_2 + FeSO_3 + 3CO_2$$

$$Fe_2(CO_3)_3 + 2SO_2 + 1/2\ O_2 \rightleftarrows 2FeSO_4 + 3CO_2$$

$$Fe_2(SO_4)_3 + 2FeO + SO_2 \rightleftarrows 4FeSO_3$$

One can, of course propose a large number of such reactions if oxygen is included as a possible reactant. The number of reactions which can be formulated which do not involve outside oxygen is much smaller. Where the reactant ferric salt is other than the hydroxide or oxide, there are problems in finding reactions which do not involve outside oxygen and in which the oxygen balance is preserved. It is for this reason that sulfite species are postulated as products in the proposed reactions. Since these reactions are thermodynamically favorable, overall reactions resulting from oxidation of some or all of the S^{+4} to S^{+6} will be still more favorable.

SECTION V - BIBLIOGRAPHY

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APPENDIX I

PROCESS REVIEWS

SURVEY OF SO_2 REMOVAL PROCESSES. INORGANIC SOLIDS

NAME: Reinluft Process

ASSIGNORS: Reinluft G. M. b. H. By F. Johswich et al.

SORPTION SECTION

Sorbent: Charcoal, peat coke or semicoke, pref ∼2 Cm particles.

Temp: 125°F <T<575.F, preferably 250.F to 300°F.

H_2O: $pSO < pH_2O <$ Saturation
O_2: $0.5\ pSO_2 < pO_2 \sim < 38mm$
CO_2: No limits
SO_2: $<10\%$ by vol.

Mode: Oxidative Sorption--Adsorption of SO_2, catalytic oxidation to SO_3, hydration of SO_3 to H_2SO_4 in micropores.

Pref. Contact: Moving Bed - Solids res. 2 hrs, gas res. 10 to 20 secs.

Thermo Chem: Highly favorable.

Eq. Sorption Data: Function of carbon structure and temperatures. Saturation of the sorbent corresponds to filling of the micropores by sorbed acid, and so occlusion of surface.

Kinetic Data: At low temp, limited by rate of oxidation of SO_2, at high temperature, limited by low surface concentration of SO_2.

REGENERATION SECTION

Temp: 570°F< T< 930°F

Reagents: SO_3 reduced by carbon in the sorbent, swept away by heated stack gas.

Byproducts: Stack gas enriched in SO_2 and water, suitable contact process feed, but the water present will make acid recovery somewhat difficult.

Pref Contactor: Moving Bed - Short solids residence time.

Thermochem: $2H_2SO_4 + C \rightarrow 2H_2O + 2SO_2 + CO_2$ has highly negative Free E. for all temps above room temp. Reaction occurs, however, only at temperatures where there is significant dissociation of H_2SO_4 to H_2O & SO_3.

Eq. Des. Data: At regen. temp. none of the product species seem to be sorbed.

Kinetic Data: Generally, regeneration is heat transfer limited.

STATUS: Several pilot and demonstration scale units have been operated. A demonstration unit at Cologne experienced problems with ignition of char in the absorber; a 10 mw unit at Luenen is operating smoothly with about 26 weeks on stream logged in one year of operation. Performance has been slightly below design. (75% SO_2 removal)

ECON. Katell estimates capital cost to be $17.77 per kw for an 800 mw installation burning 3% S coal, and operating costs to be 0.857 mills/kwh or $2.45 per ton of coal burned. Estimates for a 300 mw plant burning 1.5% S coal are $20.75/kw and 1.25 mills/kwh. Costs are exclusive of byproduct credit.

A scale exponent of 0.9 on power plant size, and 0.7 on unit absorber size would be appropriate. The above computations are based on 10 Mw units.

PROCESS DESCRIPTION

Sulfur dioxide is adsorbed from the stack gas onto a slowly moving bed of walnut-sized lumps of char. The adsorbed SO_2 is oxidized to SO_3 by the catalytic action of the carbon. This is hydrated to sulfuric acid which remains as a condensed phase within the char. The char containing adsorbed acid moves downward into a regenerator where it is heated to about 700.F with heated flue gas. At this temperature the sulfuric acid dissociates to SO_3 and water, and the sulfur trioxide reacts with carbon to form CO_2 and SO_2. The enriched SO_2 stream is fed to a sulfuric acid plant. The regenerated char is returned to the top of the adsorber after screening to remove fines. The process flows are shown in Figure 1.

Inventor suggests that off gas be fed to sulfuric converter, gases be cooled and acid recovered by a precipitator at such temperature that he gets 98% acid.

MASS BALANCE

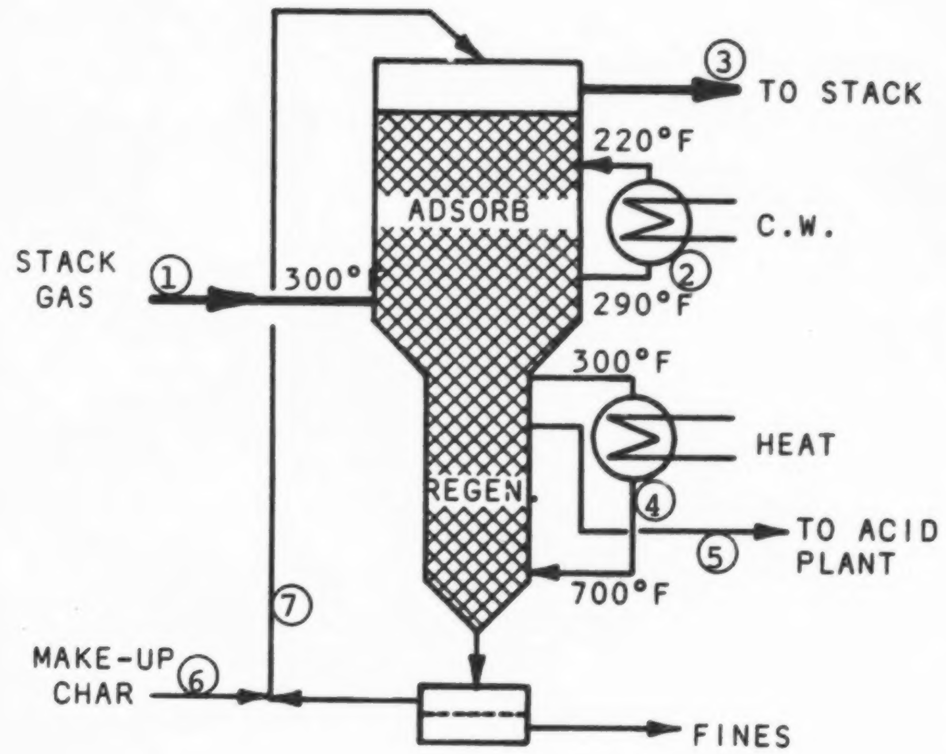
An individual 10 Mw unit would process 3 tons of char per hour, and would produce 622# of acid per hr (100% basis).

ENERGY BALANCE - Per unit absorber @ 10 Mw

Cooling water - 10M gp hr. C.W. @ 20° rise.
Heat - 1.1 therms per hr. @ up to 700°F.
Also require driver power for the recirculation and draft blowers.

Figure 3

REINLUFT PROCESS


③
TO STACK
220°F
ADSORB
C.W.
STACK GAS
①
300°
②
290°F
300°F
HEAT
REGEN.
④
TO ACID PLANT
⑤
700°F
⑦
MAKE-UP CHAR
⑥
FINES

Basis - 100 moles stack gas

	①	②	③	④	⑤	⑥	⑦
N_2	74.9	74.6	74.6	2.31	0.302	-	-
CO_2	14.7	15.4	14.5	1.48	0.194	-	-
H_2O	7.3	7.7	7.0	2.30	0.300	-	-
O_2	2.8	2.9	2.7	0.084	0.011	-	-
SO_2	0.3	0.3*	0.03	2.06	0.27	-	-
Char	-	-	-	-	-	3.63#	170#
T	300°F	290°F	215°F	300°F	700°F	100°F	687°F

*Trace SO_2 content removed.

SURVEY OF SO_2 REMOVAL PROCESSES. INORGANIC SOLIDS

NAME: "Sulfacid" Process (Pauling or Stratmann Process)

ASSIGNORS: Basic patent assigned to Metallgesellschaft A.G., Frankfurt.

SORPTION SECTION

Sorbent: Activated carbon impregnated with metals, iodine, or arsenic.

Temp: 100-160°F

H_2O: Saturated
O_2: $PO_2 < 1/2\ PSO_2$
CO_2: Any
SO_2: 10% down to stack gas levels

Mode: Oxidative sorption

Pref. Contact: Fixed bed - Concurrent downflow

Thermo Chem.: Highly favorable

Eq. Sorption Data: N.A.

Kinetic Data: Residence time in bed is 1 or more seconds for gas. Optimum temperature is a balance between rate and water vapor pressure.

REGENERATION SECTION

Byproducts: H_2SO_4, up to 20% conc. directly, or up to 85% with a precooler/evaporator. Incoming gases may be cooled and saturated with water vapor in either a countercurrent packed tower or a venturi scrubber prior to the reactor bed. The dilute acid product from the adsorber is used to cool and saturate the incoming gas. In the process, the acid is concentrated to a level fixed by the temperature and humidity of the stack gas.

STATUS

The process has been successfully applied to the treatment of chemical plant waste streams. A plant operating on up to 3000 scfm flue gas from an oil fired burner is performing well. A plant to handle 30M scfm of sulfuric acid tail gas is under construction. Small plants have operated up to 3 years. This is the most advanced of the SO_2 control processes.

ECONOMICS

Lurgi has quoted capital costs of $14.5 per kw (8000 h/a) for a 120 Mw oil fired power station. Projected operating costs are 0.4 mills/kwh. The process is well suited to small installations, but would presumably have a fairly high scale exponent, say 0.9. Acid concentration would be a problem. If incoming gas is dusty, the acid would have to be dissociated and recombined to produce a marketable byproduct. The process is particularly well suited to sulfuric acid tail gas treatment where there is no dust problem, and where the dilute acid may be substituted for a portion of the water feed to the absorber.

PROCESS DESCRIPTION

Stack gas is passed through a fixed bed of active carbon at about 160°F. The active carbon adsorbs the sulfur dioxide and catalyzes its oxidation to SO_3. This in turn reacts with water to form sulfuric acid. The acid is continuously flushed from the bed by a stream of wash water sprayed over the top. Acid of 10 to 20 percent concentration is produced. The activity of the carbon is maintained by the addition of one of several transition metals, or iodine, or arsenic.

The incoming stack gas is cooled and the acid concentrated by evaporation in a Venturi scrubber and separator. The concentration of the product acid depends strongly on the inlet gas temperature and moisture content. For a typical stack gas entering at 300°F, 83% acid is theoretically obtainable. Figure 2 shows the basic flows of this process. Alternatively, a countercurrent tower packed with coke may be used as a cooler/evaporator.

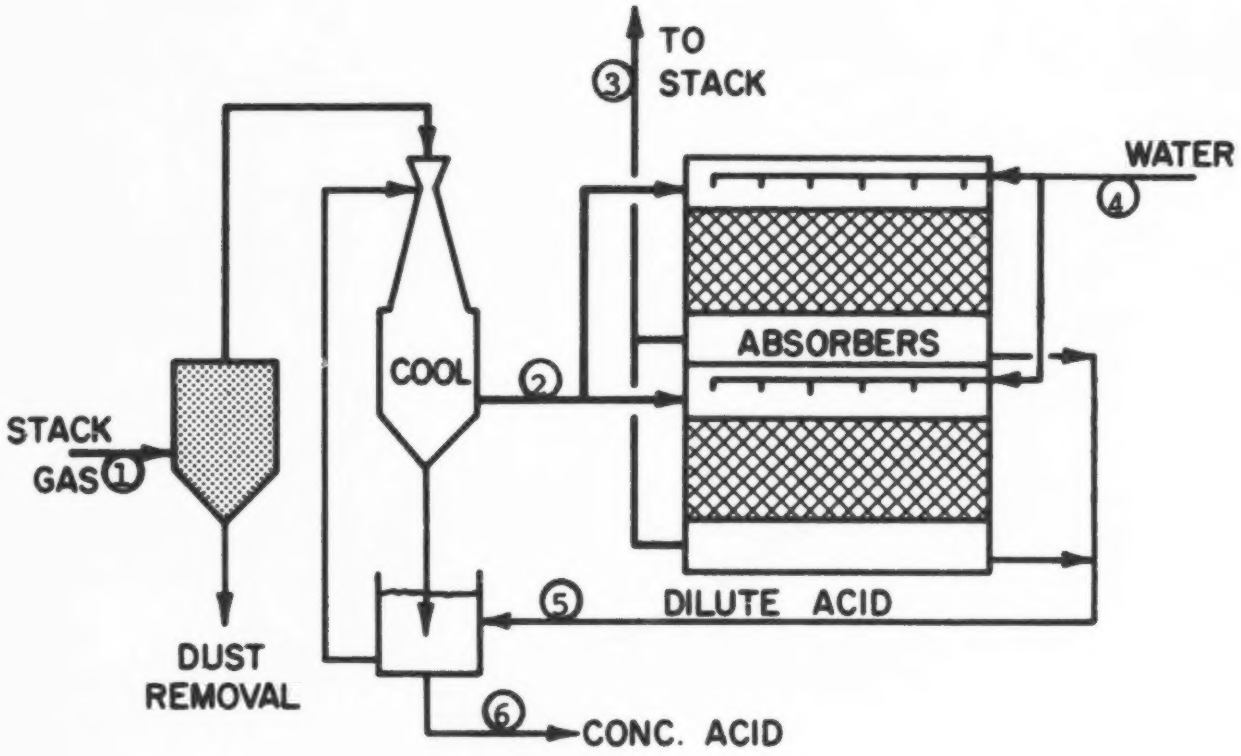
MASS AND ENERGY BALANCES

The balance shown above is one of many possible operating conditions for this process. The required inlet gas temperature is fixed by the operating temperature of the absorber. For absorber temperatures of 130 and 120°, the required inlet gas temperatures are 360°F and 250°F, respectively. Significantly less water is also required, although the product acid would probably be more dilute than the 70 wt.% shown.

COMMENTS

This process is very well suited to comparatively small waste gas streams, such as sulfuric acid tail gas. The process is the most advanced of those available. Operation is very simple. The corrosion conditions are severe. The high specific water consumption and the cold, wet plume are potential problem areas. Plume reheat would be required for large scale installations.

"SULFACID" PROCESS


TO STACK
③
WATER
④
COOL
②
ABSORBERS
STACK GAS ①
⑤ DILUTE ACID
DUST REMOVAL
⑥ CONC. ACID

	①	②	③	④	⑤	⑥
N_2	74.9	74.9	74.9			
CO_2	14.7	14.7	14.7			
H_2O	7.3	18.3	22.6	16.5	11.9	0.63
O_2	2.8	2.8	2.67			
SO_2	0.3	0.3	0.03			
H_2SO_4	–	–	–		0.27	0.27
T	510°F	230°F	140°F	–	140°F	230°F
Σwt.	2984#		3238#	297#	240#	37.7#

In pound moles unless indicated.

Figure 4

SURVEY OF SO_2 REMOVAL PROCESSES. INORGANIC SOLIDS

NAME: Hitachi Process

ASSIGNORS: Hitachi LTD.

SORPTION SECTION

Sorbent:	Activated carbon - not consumed - moderate to large briquettes or particles.
Temp.:	$\sim$300 to 200°F inlet, 55°F lower outlet
H_2O:	Oil or coal flue gases are satisfactory
O_2:	As Reinluft
CO_2:	No limits
SO_2:	As Reinluft
Mode:	As Reinluft
Pref. Contact:	Fixed bed
Thermo Chem.	Highly favorable
Eq. Sorption Data:	No data - not an equilibrium situation since sorbed material is converted to capillary condensed phase.
Kinetic Data:	Unknown

REGENERATION SECTION

Temp.:	<212°F for wash, incoming stack temp. for drying.
Reagents:	Water, air
Byproducts:	Dilute sulfuric acid (10-20%)
Pref. Contactor:	Fixed beds
Eq. Des. Data:	Only a portion of gas used to dry the towers so that effluent gas is never fully saturated with water.
Kinetic Data:	Desorption is diffusion controlled, preferably operated with several towers in a counter-current manner.

STATUS

A 2 Mw equivalent plant is operating at Tokyo Electric's Gai plant. A 50 Mw unit is under design.

ECONOMICS

$10/kw capacity @ 175 Mw, 90% removal (assume 8000 hrs)
0.45 mills/kwh exclusive of byproduct. Scale factor would be expected to be near 1.0, say 0.9

PROCESS DESCRIPTION

Stack gas passes first to an electrostatic precipitator where dust is removed. Then it goes to a flow splitter where a portion is led to a tower containing water saturated carbon. The gas leaves the tower cool and water-saturated. Pressure is boosted with a draft fan and the gas mixes with the rest of the incoming gas. The combined stream at about 245°F passes through a fixed bed of dry active carbon. The sulfur dioxide is adsorbed, oxidized, and converted to sulfuric acid sorbed within the carbon structure. The gas is discharged at substantially the same temperature as it entered the absorber. When a given adsorbing tower approaches saturation, the gas flows are diverted, and water is sprayed over the bed to wash the sulfuric acid out of the carbon. To increase the concentration of the product acid, several towers may be extracted countercurrently. Twenty percent is about the most concentrated acid achievable with this system. A typical cycle calls for 30 hrs adsorb, 10 hrs wash, and 20 hrs dry.

ENERGY BALANCE

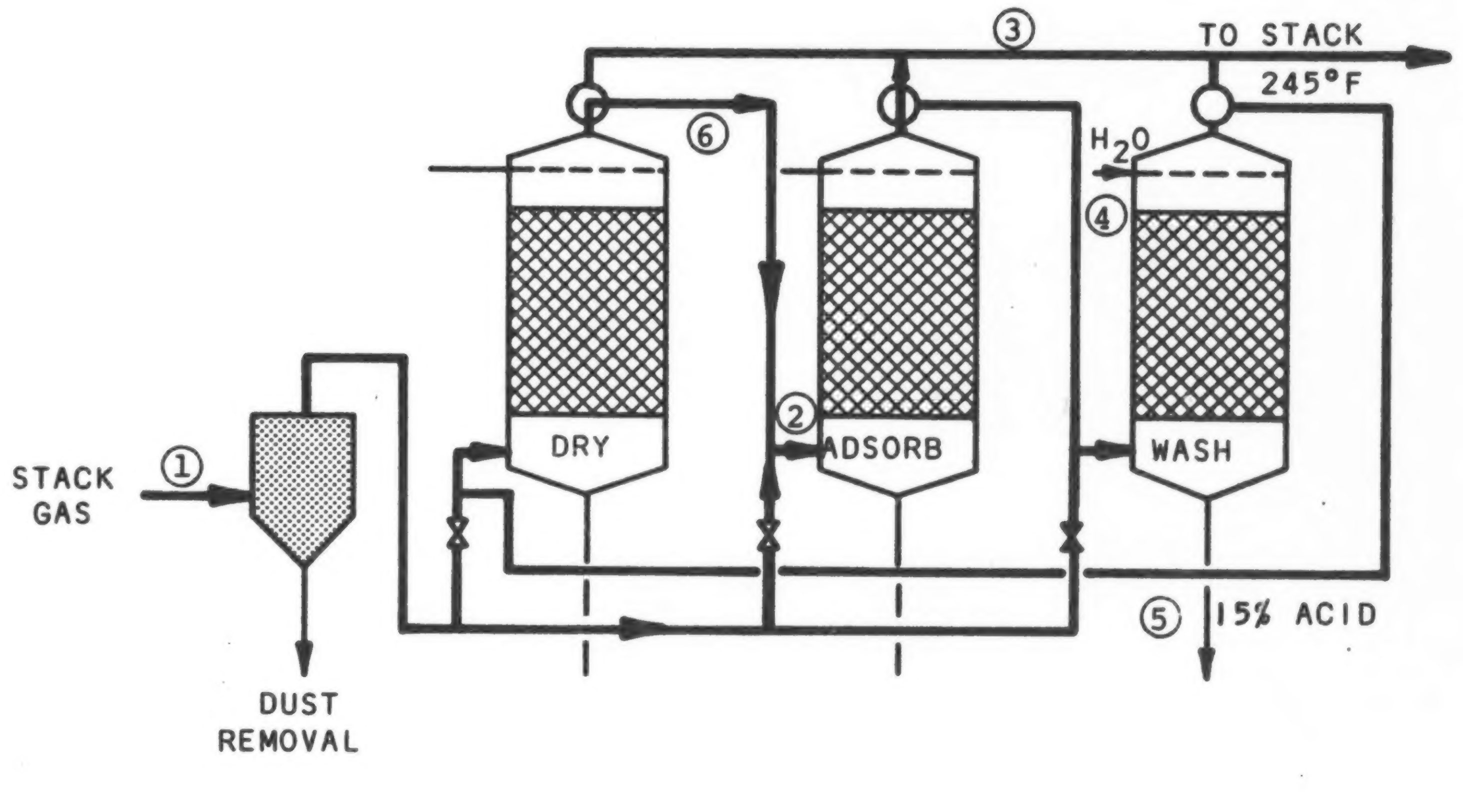
No net energy input is required by the process as shown, except drives for draft fans. Steam would also be required to concentrate the dilute product acid in any actual installation.

COMMENTS

The Hitachi process has as its major problem the dilute byproduct acid produced by evaporation. To have any byproduct value, this product would have to be upgraded to ∿70% acid. The concentrated product is useful for only limited uses, and might require dissociation and recombination to produce a product of saleable value. U.S. conditions do not make such reprocessing of acid favorable at present sulfur prices. Because of the unfavorable scale exponent, very large process installations would have little economic advantage.

Figure 5

Hitachi Process


③
TO STACK
245°F
⑥
H_2O
④
STACK GAS
①
DRY
②
ADSORB
WASH
⑤
15% ACID
DUST REMOVAL

Basis - 100 moles of gas in.

	①	②	③	④	⑤	⑥
N_2	79.4	74.9	74.9			21.7
CO_2	14.7	14.7	14.7			4.3
H_2O	7.3	9.6	9.3	191.4#	150#	4.1
O_2	2.8	2.8	2.67			0.81
SO_2	0.3	0.3	0.03			0.1
H_2SO_4	-	-	-	-	26.46#	-
Wt.	2984.2#	3025.6#	2999.1#	191.4#	176.5#	901.1
T	300°F	245°F	245°F	-		125°F

SURVEY OF SO_2 REMOVAL PROCESSES. INORGANIC SOLIDS

NAME: Grillo Process

ASSIGNORS: A.G. fur Zinkindustrie vorm Wilhelm Grillo

SORPTION SECTION

Sorbent: Mixture of transition metal oxide and alkaline earth hydroxide, pref $MnO_2 + Mg(OH)_2$

Temp.: Presumed to be 200 to 250°F

H_2O: Less than saturation
O_2: None required
CO_2: Some slight interference with chemistry
SO_2: Any

Mode: Reaction of SO_3 with MnO_2 and with $Mg(OH)_2$ Gas phase reaction presumably with $Mg(OH)_2$ with transfer in slurry to MnO_2 forming $MnSO_4$

Pref. Contact: Spray Tower

Thermo Chem.: Highly favorable for reaction

Eq. Sorption Data: Probable limit on the per pass SO_2 pickup set by the $Mg(OH)_2$ content.

Kinetic Data: Unknown

REGENERATION SECTION

Temp.: Unknown, presumably 1500°F

Reagents: Air, coal for fuel and possibly for reductant.

Byproducts: High SO_2 stream, presumably converted to sulfuric acid.

Pref. Contactor: Multiple hearth furnace

Thermo Chem.: To be established.

Kinetic Data: Unknown

STATUS

Small scale pilot plant tests have been completed by A.G. fur Zinkindustrie vorm Wilhelm Grillo. An 8 Mw unit constructed near Cologne and went into operation in October 1967. This is to handle about 12,000 scfm of oil burner stack gas or sulfuric acid tail gas.

ECONOMICS

No economics have been published, to our knowledge, on the process. The process conditions are not sufficiently well known to be able to make an intelligent cost estimate. The number of processing steps and the number of mechanical operations required suggest that the process will be very expensive unless significant breakthroughs are made.

PROCESS DESCRIPTION

The Grillo process absorbs SO_2 in a mixture of a metal oxide and an alkaline earth hydroxide [$MnO_2 + Mg(OH)_2$ is preferred]. A slurry of the absorbent is sprayed into a spray dryer where it dries as it contacts the stack gas. The particles of absorbent are collected, and most are reslurried and sent back to the spray nozzle. Two spray towers are used. Fresh absorbent is fed to the slurry tank of the second tower. A small quantity of absorbent passes to the first tower each pass, and a like amount of material is removed from the first tower for regeneration. The loaded acceptor ($\sim$20% sulfur) is mixed with coal, roasted to give SO_2 suitable for acid plant feed. The calcine is slurried in water, passed through a clarifier where fly ash separates as an underflow, filtered, and dried. This is then returned to the absorbers. Figure 4 is a process flow sheet.

MASS BALANCE

At the moment we have insufficient information on either the chemistry or process conditions to develop a detailed flow sheet. We have developed a hypothetical flow sheet and mass balance, however, which is subject to certain assumptions.

ASSUMPTIONS

The following assumptions were made in deriving the material balances:

1:1 mole ratio Mn to Mg.

Complete conversion of Mn, 50% conversion of Mg (Bal.ash).

20% sulfur content in spent absorbent (Brocke).

Top calciner temp. is 1500°F ($MnSO_4$ decomp. temp.).

Coal provides fuel value only at 12,000 Btu/lb, 10% ash, comp. $(CH)_n$

Gases leave the calciner at 1000°F, 10% X's air.

Solids leave calciner at 1500°F.

A 2% slurry is fed to clarifier, separation is perfect, bottoms are 5% ash.

Filter cake is 40% solids.

Recycle slurry to dryers is 30% solids.

Mean recycle ratio for absorbers in combination is 10:1.

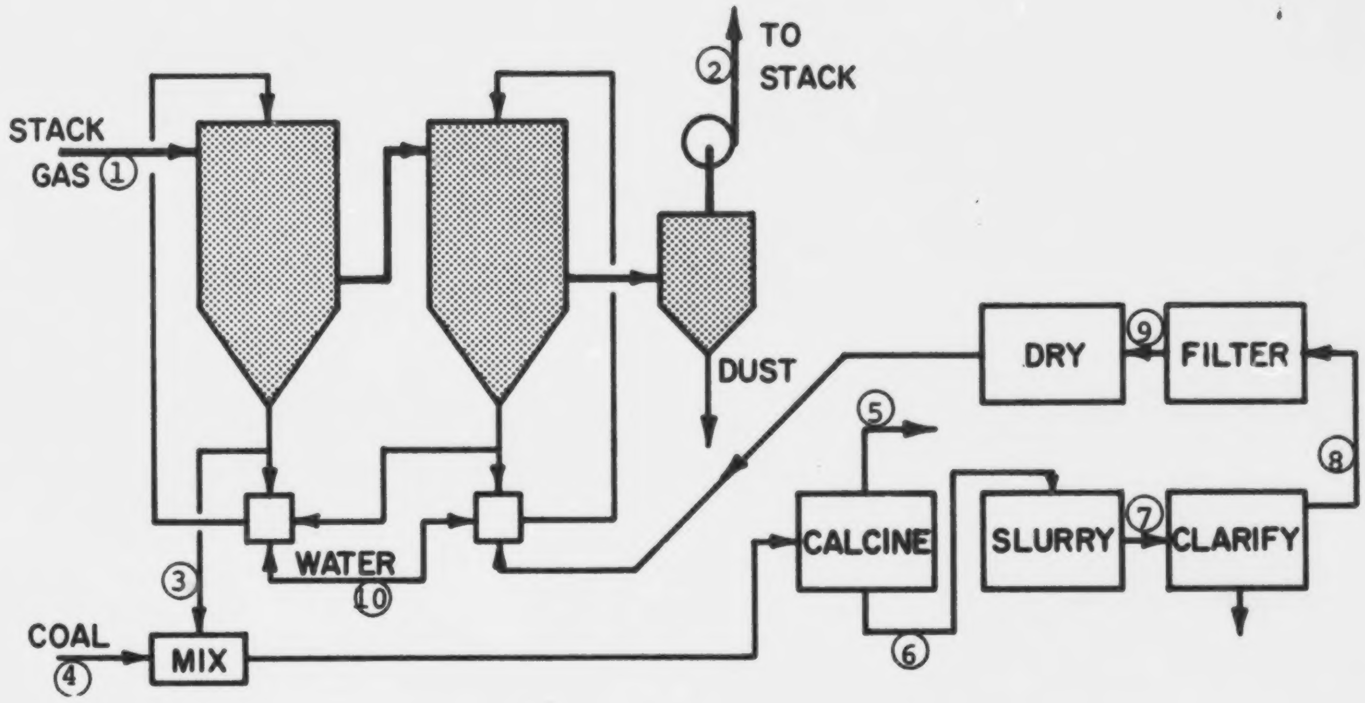
The roasting operation can be carried out in such a fashion as to produce SO_2 directly without first completely reducing all sulfur species to COS, S, and H_2S.

ENERGY BALANCE

0.174 lbs coal per lb SO_2 are required for regeneration. There will also be a large driver requirement for a booster fan and for machinery drives.

Figure 6

GRILLO PROCESS


STACK GAS ①
② TO STACK
DUST
DRY
⑨
FILTER
⑤
⑧
CALCINE
SLURRY
⑦
CLARIFY
③
WATER
⑩
COAL
④
MIX
⑥

Basis - 100# moles of stack gas

	①	②	③	④	⑤	⑥	⑦	⑧	⑨	⑩
N_2	74.9	74.9			1.369					
CO_2	14.7	14.7			0.208					
H_2O	7.3	20.9			0.171					
O_2	2.8	2.8			0.011					
SO_2	0.30	0.03			0.27				36.74	
Water		(sat.)					1294.8	1258.3		244
MnO_2			–			0.169	0.169	0.169	0.169	
$MnSO_4$			0.169							
$MgSO_3$			0.101							
MgO			–			0.169				
$Mg(OH)_2$			0.068				0.169	0.169	0.169	
Coal				3.0#						
Ash			1.63#	0.3#		1.93#	1.93#	–		
T	340°F	120°F	RT	RT	1000°F	1500°F	RT	RT	RT	RT
ΣWt.	2984	3210.7					1321.4		61.24	367

SURVEY OF SO_2 REMOVAL PROCESSES. INORGANIC SOLIDS

NAME: Still Process

ASSIGNORS: Firma Karl Still

SORPTION SECTION

Sorbent:	Hydrated basic lignite ash (possibly augmented or supplanted by hydrated lime).
Temp.:	850°F to 200°F.
H_2O:	Depends on temperature, >P decomp. $Ca(OH)_2$
O_2:	Any
CO_2:	No interference noted in literature.
SO_2:	Any
Mode:	Reaction of calcium hydroxide with SO_2.
Pref. Contact:	Transport bed.
Thermo Chem.:	Highly favorable. Temperature limited by decomposition of $Ca(OH)_2$.
Kinetic Data:	Unknown. Reaction is probably controlled by diffusion of gas into the reacted solid.

REGENERATION SECTION

Partial regeneration may be practiced, or spent absorbent may be discarded.

Reagents:	Inert gas sweep.
Byproducts:	SO_2, $CaSO_4$
Pref. Contactor:	Kiln or multiple hearth
Thermo Chem.:	Multiple favorable reaction paths, thermal dissociation of $CaSO_3$ can go to CaO and SO_2, or to CaS and $CaSO_4$.
Kinetic Data:	Unknown.

STATUS

Small scale demonstration was accomplished by Firma Karl Still. A 10 Mw unit was to have started operation at Herne Power Station near Recklinghausen, Germany, November, 1967.

ECONOMICS

Capital costs of $5.00/kwh and 0.5 mills/kwh operating expense are claimed by Still, for a 200 Mw installation. Value assigned to ash is unclear.

PROCESS DESCRIPTION

Gases from the power plant dust collector are contacted with a hydrated lignite ash in sequential transport contactors. The ash is collected by cyclones and bag filters, and the off gases are led to a stack. The absorbent is regenerated by heating. During regeneration an SO_2 rich stream is produced which may be fed to a sulfuric acid plant. The absorbent is then hydrated before being returned to the absorber.

Only the ash from lignites with a high lime content are suitable as absorbents. Unfortunately, with repeated cycling, more and more of the calcium in the sorbent is bound irreversibly as calcium sulfate. Thus, the lignite ash must be inexpensive and readily available. The flows are shown in Figure . Efforts will be made to operate the pilot plant with hydrated lime.

MASS AND ENERGY BALANCE

The specific performance of the Still process will be strongly dependent on the make-up of the ash. Minor constituents are likely to determine the quantities of $CaSO_4$ formed during absorption and regeneration.

Assuming 90% SO_2 removal, and based on 100 moles of entering stack gas, the process will use 9.7# of water. The calciner will consume about one pound of coal per ten to fifteen pounds of solids throughput. The magnitude of the solids purge is likely to be considerable (say, 20-25%) under the best conditions, but under less favorable conditions, even more material may have to be purged.

MASS BALANCE

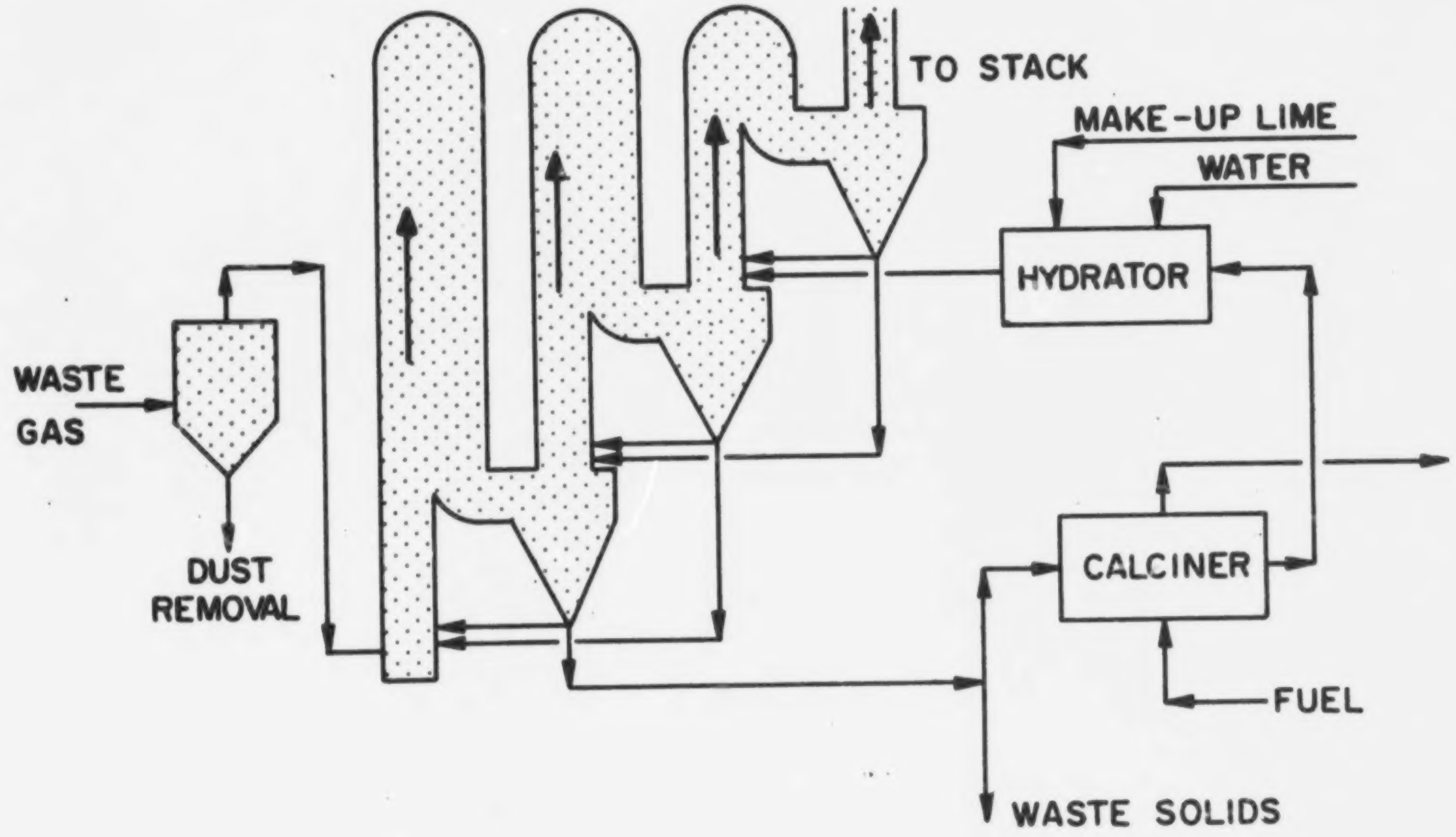
Insufficient data are available to specify the recycle ratios required for various absorbents. Regeneration, too, will depend strongly on minor constituents in the ash.

ENERGY BALANCE

Calciner fuel is the only extraneous energy requirement.

FIGURE 7

STILL PROCESS


TO STACK
MAKE-UP LIME
WATER
HYDRATOR
WASTE
GAS
DUST
REMOVAL
CALCINER
FUEL
WASTE SOLIDS

APPENDIX II

THERMODYNAMIC COMPUTATIONS

TABLE XVI

THERMODYNAMIC VALUES FOR MISCELLANEOUS COMPOUNDS

	H°	S°	A	B	C	D
H_2SO_4	-194.548	37.501	36.77	9.16 -3	-2.088-6	-5.432 +5
SO_3	-94.470	61.345	9.054	1.679-2	-7.659-6	-11.3429+4
SO_2	-70.947	59.297	7.0756	1.076-3	-4.848-6	-2.9107+4
S_2	+30.84	54.51	7.866	1.892-3	8.996-7	-2.9053+4
CO_2	-94.035	51.125	6.94	9.742-3	3.667-6	-5.7464+4
CO	-26.416	41.218	5.735	2.703-3	-5.356-7	4.2111+4
C	0	1.372	-0.221	9.963-3	-4.611-6	-2.7007+4
O_2	0	49.007	5.179	5.24 -3	2.141-6	4.186+4
H_2	0	31.208	7.567	-1.5 -3	1.193-6	-2.9332+4
H_2O	-57.798	45.105	6.954	2.569-3	3.152-7	2.4706+4
SiO_2	-217.5	10.0	-5.364	5.413-2	-3.396-5	2.57565+5

Note: Positive and negative integers after values in Columns B, C, and D are exponents of powers of 10. Thus, 9.16-3 is 9.16×10^{-3}.

TABLE XVII - REGENERATION REACTIONS

TABULATIONS OF Kp FOR VARIOUS REACTIONS

$SO_3 = 1/2\ O_2 + SO_2$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	16827.9	-12.3424	4.54558 E-13
400	14514.5	-7.93503	1.16137 E-8
500	12185.	-5.32917	4.68633 E-6
600	9816.83	-3.57788	2.64317 E-4
700	7418.	-2.31736	4.81546 E-3
800	4991.96	-1.36454	4.31976 E-2
900	2540.16	-.617198	.241436
1000	62.9723	-1.37707 E-2	.968789

$H_2SO_4 = SO_3 + H_2O$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	21722.9	-15.9326	1.16783 E-16
400	15733.2	-8.60126	2.50461 E-9
500	10250.1	-4.48293	3.28904 E-5
600	5135.54	-1.87171	1.34365 E-2
700	356.905	-.111496	.773578
800	-4114.37	1.12465	13.3246
900	-8301.43	2.01704	104.003
1000	-12222.6	2.67282	470.786

$2H_2SO_4 + C = 2\ SO_2 + CO_2 + 2\ H_2O$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	-17155.9	12.583	3.82829 E 12
400	-34101.9	18.6434	4.39900 E 18
500	-50099.8	21.9114	8.15504 E 21
600	-65480.8	23.8653	7.33401 E 23
700	-80296.2	25.0843	1.21424 E 25
800	-94597.	25.8579	7.20881 E 25
900	-108428.	26.3453	2.21441 E 26
1000	-121825.	26.6405	4.36988 E 26

TABLE XVII (Continued)

$H_2SO_4 + C = H_2O + SO_2 + CO$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	7560.38	-5.54516	2.84997 E-6
400	-2625.83	1.43553	27.2603
500	-12410.6	5.42785	267824.
600	-21966.9	8.00612	1.01419 E 8
700	-31309.8	9.78109	6.04067 E 9
800	-40459.7	11.0596	1.14697 E 11
900	-49436.4	12.0118	1.02763 E 12
1000	-58257.8	12.7397	5.49162 E 12

$H_2SO_4 + CO = H_2O + SO_2 + CO_2$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	-24716.3	18.1282	1.34327 E 18
400	-31476.1	17.2078	1.61371 E 17
500	-37689.2	16.4836	3.04492 E 16
600	-43513.9	15.8592	7.23139 E 15
700	-48986.4	15.3032	2.01011 E 15
800	-54137.4	14.7983	6.28507 E 14
900	-58991.2	14.3334	2.15487 E 14
1000	-63567.3	13.9008	7.95735 E 13

$H_2SO_4 + H_2 = 2\ H_2O + SO_2$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	-16084.9	11.7975	6.27307 E 11
400	-23185.5	12.6754	4.73596 E 12
500	-29748.	13.0104	1.02435 E 13
600	-35922.5	13.0924	1.23714 E 13
700	-41742.5	13.0402	1.09709 E 13
800	-47239.9	12.9129	8.18299 E 12
900	-52441.6	12.742	5.52123 E 12
1000	-57370.	12.5455	3.51195 E 12

TABLE XVII (Continued)

$SO_3 + C = CO + SO_2$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	-14162.5	10.3875	2.44039 E 10
400	-18359.	10.0368	1.08840 E 10
500	-22660.7	9.91078	8.14292 E 9
600	-27102.4	9.87783	7.54804 E 9
700	-31666.7	9.89258	7.80874 E 9
800	-36345.3	9.9349	8.60796 E 9
900	-41135.	9.99479	9.88081 E 9
1000	-46035.2	10.0669	1.16648 E 10

$SO_3 + CO = SO_2 + CO_2$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	-46439.2	34.0608	1.15022 E 34
400	-47209.3	25.8091	6.44293 E 25
500	-47939.2	20.9665	9.25779 E 20
600	-48649.4	17.7309	5.38191 E 17
700	-49343.3	15.4147	2.59845 E 15
800	-50023.	13.6737	4.71691 E 13
900	-50689.8	12.3164	2.07193 E 12
1000	-51344.7	11.2279	1.69023 E 11

$SO_3 + H_2 = H_2O + SO_2$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	-37807.8	27.7301	5.37154 E 27
400	-38918.7	21.2767	1.89089 E 21
500	-39998.	17.4934	3.11443 E 17
600	-41058.	14.9641	9.20730 E 14
700	-42099.4	13.1517	1.41820 E 13
800	-43125.5	11.7883	6.14128 E 11
900	-44140.2	10.725	5.30873 E 10
1000	-45147.3	9.87272	7.45975 E 9

$2\ SO_3 + C = 2\ SO_2 + CO_2$

T[K]	DEL F[CALS]	LOG10 K	K EQ
298.15	-60601.6	44.4482	2.80700 E 44
400	-65568.3	35.8459	7.01251 E 35
500	-70599.9	30.8773	7.53854 E 30
600	-75751.9	27.6088	4.06229 E 27
700	-81010.	25.3073	2.02907 E 25
800	-86368.3	23.6086	4.06029 E 23
900	-91824.8	22.3112	2.04724 E 22
1000	-97379.8	21.2948	1.97161 E 21

TABLE XVIII

THERMODYNAMIC VALUES FOR BARIUM COMPOUNDS

			Cp (T)			
	H_f°	S_f°	A	B	C	D
BaO_1	-132.07	16.795	12.733	1.0392-3		1.9829+5
$Ba()_2H)_2$	-226.02	(12.613)	16.90	21.90 -3		
$BaCO_3$	-287.16	26.781	20.76	1.1694-2		4.7708+5
$BaSO_3$	-282.6	28.60	19.64	1.614 -2		1.98 +5
$BaSO_4$	-349.99	31.487	33.759	3.401 -5		8.394 +5
BaS	-106.0	19.52				
$BaSiO_3$	-359.5					
$Ba_3(PI_4)_2$	-998.0					
$BaHPO_4$	-465.8					

Note: Positive and negative integers after values in Columns Band D are exponents of powers of 10. Thus,9.16-3 is 9.16 x 10^{-3}.

TABLE XIX

REACTIONS OF BARIUM COMPOUNDS

$$BaCO_3 + SO_2 \rightleftarrows BaSO_3 + CO_2$$

```
T(K)             DEL F(CALS)     LOG10 K          K EQ
 298.15          -16633.9         12.2001          1.58523E+12
 400             -16140.2         8.82378         666461819
 500             -15553.7         6.80251         6.34621E+06
 600             -14933.3         5.44264         277099.
 700             -14313.6         4.47154         29616.8
 800             -13717.3         3.74959         5618.05
 900             -13161.1         3.19783         1577.
 1000            -12659.1         2.76826         586.491
```

$$Ba(OH)_2 + SO_2 \rightleftarrows BaSO_3 + H_2O$$

```
T(K)             DEL F(CALS)     LOG10 K          K EQ
 298.15          -43966.2         32.247           1.76594E+32
 400             -44407.6         24.2774          1.89410E+24
 500             -45013.9         19.6871          4.86526E+19
 600             -45740.8         16.6708          4.68628E+16
 700             -46566.4         14.5472          3.52542E+14
 800             -47480.7         12.9787          9.52213E+12
 900             -48479.1         11.7792          6.01513E+11
 1000            -49560.2         10.8377          6.88210E+10
```

$$Ba(OH)_2 + 1/2\ O_2 + SO_2 \rightleftarrows BaSO_4 + H_2O$$

```
T(K)             DEL F(CALS)     LOG10 K          K EQ

OVERFLOW   IN 580
 298.15          -104911.         76.9471          5.78960E+76
 400             -103267.         56.4553          2.85330E+56
 500             -102325.         44.7525          5.65587E+44
 600             -101834.         37.1149          1.30278E+37
 700             -101688.         31.7671          5.84864E+31
 800             -101825.         27.8337          6.81791E+27
 900             -102207.         24.8338          6.82003E+24
 1000            -102807.         22.4817          3.03176E+22
```

$$BaCO_3 + 1/2\ O_2 + SO_2 \rightleftarrows BaSO_4 + CO_2$$

```
T(K)             DEL F(CALS)     LOG10 K          K EQ
 298.15          -77578.9         56.9002          7.94704E+56
 400             -74999.3         41.0017          1.00397E+41
 500             -72864.9         31.8679          7.37748E+31
 600             -71026.8         25.8867          7.70333E+25
 700             -69435.3         21.6914          4.91339E+21
 800             -68061.7         18.6045          4.02257E+18
 900             -66888.9         16.2524          1.78802E+16
 1000            -65906.2         14.4122          2.58366E+14
```

TABLE XX

THERMODYNAMIC VALUES FOR CALCIUM COMPOUNDS

CALCIUM

	H°	S°	Cp(T)			
			A	B	C	D
MO	-151,730	9.484	11.857	1.0798-3	0	166,040
$M(OH)_2$	-235,600	19.924	25.131	2.8822-3	0	450,780
MCO_3	-288,110	22.194	24.965	5.237-3	0	619,700
MSO_3	-275,880	24.201	18.770	1.618-2	0	166,000
MSO_4	-240,190	25.491	17.220	2.337-2	0	32,560
MS	-115,300	13.500	10.20	3.80-3	0	0
$MSiO_3$	-377,400	20.900	25.85	3.94-3	0	565,000
$M_3(PO_4)_2$	-435,200	21.0				

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus,9.16-3 is 9.16 x 10^{-3}.

TABLE XXI

REACTIONS OF CALCIUM COMPOUNDS

$CaCO_3 + SO_2 \rightarrow CaSO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-9019.91	6.61565	4.12711E+06
400	-8417.59	4.60185	39981.1
500	-7535.64	3.29576	1975.86
600	-6488.44	2.3648	231.633
700	-5340.43	1.66833	46.5945
800	-4132.57	1.12963	13.4781
900	-2894.37	.703262	5.04966
1000	-1648.93	.360585	2.29395

$Ca(OH)_2 + SO_2 \rightarrow CaSO_3 + H_2O$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-24174.8	17.731	5.38304E+17
400	-23114.3	12.6365	4.32989E+12
500	-21850.1	9.55625	3.59960E+09
600	-20442.6	7.45058	2.82214E+07
700	-18935.6	5.91542	823037.
800	-17358.	4.74477	55561.
900	-15731.4	3.82233	6642.54
1000	-14072.6	3.07736	1194.98

$CaSiO_3 + SO_2 \rightarrow CaSO_3 + SiO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-31319.3	22.9711	9.35667E+22
400	-24685.3	13.4953	3.12844E+13
500	-17517.8	7.6615	4.58674E+07
600	-9792.94	3.56917	3708.22
700	-1555.88	.486051	3.06232
800	7171.46	-1.9603	1.09572E-02
900	16380.8	-3.98013	1.04681E-04
1000	26072.6	-5.7015	1.98839E-06

$CaCO_3 + 1/2\ O_2 + SO_2 \rightarrow CaSO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-66408.8	48.7075	5.09925E+48
400	-63368.4	34.6432	4.39745E+34
500	-59930.4	26.2109	1.62521E+26
600	-56212.7	20.4875	3.07223E+20
700	-52303.7	16.3395	2.18535E+16
800	-48258.8	13.1914	1.55394E+13
900	-44117.1	10.7194	5.24051E+10
1000	-39908.1	8.72702	533357344

TABLE XXI (Continued)

$Ca(OH)_2 + 1/2\ O_2 + SO_2 \rightarrow CaSO_4 + H_2O$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-81563.7	59.8229	6.65101E+59
400	-78065.1	42.6778	4.76236E+42
500	-74244.8	32.4714	2.96080E+32
600	-70166.8	25.5732	3.74310E+25
700	-65898.8	20.5866	3.86016E+20
800	-61484.3	16.8066	6.40583E+16
900	-56954.1	13.8384	6.89360E+13
1000	-52331.7	11.4438	2.77840E+11

$CaSiO_3 + SO_2 + 1/2\ O_2 \rightarrow CaSO_4 + SiO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-88708.2	65.063	1.15606E+65
400	-79636.1	43.5367	3.44091E+43
500	-69912.5	30.5767	3.77275E+30
600	-59517.2	21.6918	4.91835E+21
700	-48519.1	15.1572	1.43627E+15
800	-36954.8	10.1015	1.26329E+10
900	-24841.9	6.03598	1.08638E+06
1000	-12186.6	2.66494	462.313

$CaS + 2\ SO_2 \rightarrow CaSO_4 + S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-36624.5	26.8622	7.28121E+26
400	-31052.9	16.9764	9.47201E+16
500	-25761.8	11.2671	1.84968E+11
600	-20596.7	7.50673	32116345
700	-15540.5	4.85482	71583.9
800	-10589.8	2.8947	784.699
900	-5746.93	1.39636	24.9094
1000	-1017.25	.22245	1.66897

TABLE XXII

THERMODYNAMIC VALUES FOR COBALT COMPOUNDS

Co(+2)	H_f°	S_f°	Cp(T)			
			A	B	C	D
CoO_1	-57.073	12.655	11.534	2.0402-3		-3.9896+4
$Co(OH)_2$	-131.2					
$CoCO_3$	-173.3	21.99	17.17	1.875 -2		9.9 +4
$CoSO_3$	-161.75	25.5	18.44	1.714 -2		-4.0 +4
$CoSO_4$	-207.34	27.067	30.095	9.915 -3		6.915 +2
CoS	-22.76	16.18	10.5952	+2.51 -3		
Co(+3)						
$Co_3)_4$	-216.2	24.607	30.818	1.7072-2		5.7575+5
$Co(OH)_3$	-176.6					

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16 x 10^{-3}.

TABLE XXIII

REACTIONS OF COABLT COMPOUNDS

$CoCO_3 + SO_2 \rightarrow CoSO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-10148.	7.44306	2.77373E+07
400	-9928.4	5.42781	267798.
500	-9719.42	4.25085	17817.5
600	-9547.96	3.47988	3019.14
700	-9430.65	2.94611	883.301
800	-9379.25	2.56379	366.263
900	-9403.54	2.28483	192.677
1000	-9512.44	2.08016	120.271

$CoCO_3 + SO_2 + 1/2\ O_2 \rightarrow CoSO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-48899.5	35.8653	7.33368E+35
400	-46766.3	25.5669	3.68914E+25
500	-44912.5	19.6428	4.39309E+19
600	-43276.7	15.7728	5.92635E+15
700	-41842.4	13.0714	1.17878E+13
800	-40597.5	11.0972	1.25091E+11
900	-39534.2	9.60584	4.03497E+09
1000	-38647.3	8.45132	2.82697E+08

$2\ SO_2 + CoS \rightleftarrows CoSO_4 + S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	4014.69	-2.94457	1.13614E-03
400	9106.22	-4.97832	1.05118E-05
500	13617.8	-5.95585	1.10702E-06
600	17757.7	-6.47202	3.37273E-07
700	21582.5	-6.74231	1.81004E-07
800	25124.9	-6.86783	1.35573E-07
900	28404.4	-6.90156	1.25440E-07
1000	31432.5	-6.8736	1.33781E-07

$SO_2 + 2\ CoS \rightarrow 2\ CoO + 3/2\ S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-10148.	7.44306	2.77373E+07
400	-9928.4	5.42781	267798.
500	-9719.42	4.25085	17817.5
600	-9547.96	3.47988	3019.14
700	-9430.65	2.94611	883.301
800	-9379.25	2.56379	366.263
900	-9403.54	2.28483	192.677
1000	-9512.44	2.08016	120.271

TABLE XXIV

THERMODYNAMIC VALUES OF COPPER COMPOUNDS

			Cp(T)			
Cu(+2)	H°_f	S°_f	A	B	C	D
CuO	-39.49	10.187	9.267	4.797-3		
$Cu(OH)_2$	-107.2					
$CuCO_3$	-142.10	21.023	14.91	2.151-2		1.39+5
$CuSO_3$	-127.28	25.70	16.18	1.99 -2		
$CuSO_4$	-184.22	27.067	18.805	1.718-2		2.792+3
CuS	-11.6	15.9	10.60	2.64 -3		
Cu(+1)						
Cu_2O	-40.78	22.521	14.893	5.698-3		
Cu_2CO_3	-142.29	37.63	20.53	2.241-2		1.39 +5
Cu_2SO_3	-127.06	40.60	21.8	2.08 -2		
Cu_2SO_4	-179.51	38.463	24.19	23.40 -2		5.8 +4
Cu_2S (103)	-19.931	28.883	19.492			
Cu_2S (350)			23.24			
			20.31			

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus,9.16-3 is 9.16 x 10^{-3}.

TABLE XXV

REACTIONS OF COPPER COMPOUNDS

$CuCO_3 + SO_2 \rightleftarrows CuSO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-7225.97	5.29988	199472.
400	-7125.2	3.89531	7858.01
500	-7032.92	3.07589	1190.94
600	-6978.16	2.54328	349.369
700	-6977.55	2.17977	151.275
800	-7042.85	1.92514	84.1675
900	-7183.84	1.7455	55.654
1000	-7409.44	1.62028	41.714

$CuS + 2\,SO_2 \rightleftarrows CuSO_4 + S_2$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	15891.2	-11.6554	2.21108E-12
400	21105.2	-11.5381	2.89664E-12
500	26121.9	-11.4246	3.76205E-12
600	31041.7	-11.3136	4.85791E-12
700	35862.7	-11.2034	6.26046E-12
800	40580.1	-11.0925	8.08239E-12
900	45187.4	-10.9794	1.04851E-11
1000	49677.1	-10.8633	1.36999E-11

$CuCO_3 + SO_2 + 1/2\,O_2 \rightleftarrows CuSO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-57267.8	42.0031	1.00706E+42
400	-54987.	30.0611	1.15106E+30
500	-52744.6	23.0682	1.17000E+23
600	-50532.2	18.4171	2.61293E+18
700	-48366.3	15.1095	1.28676E+15
800	-46258.3	12.6446	4.41150E+12
900	-44217.1	10.7437	5.54215E+10
1000	-42250.8	9.23932	1.73510E+09

TABLE XXV (Continued)

$2\ CuS + SO_2 \rightleftarrows 2\ CuO + 3/2\ S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	100662.	-73.8306	1.47720E-74
400	100410.	-54.8936	1.27765E-55
500	100326.	-43.8784	1.32327E-44
600	100360.	-36.5774	2.64633E-37
700	100478.	-31.3889	4.08426E-32
800	100656.	-27.5142	3.06083E-28
900	100877.	-24.5105	3.08650E-25
1000	101122.	-22.1131	7.70770E-23

$Cu_2CO_3 + SO_2 \rightleftarrows Cu_2SO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-6307.02	4.62588	42255.6
400	-6032.4	3.29788	1985.57
500	-5769.42	2.52329	333.65
600	-5543.96	2.02057	104.851
700	-5372.65	1.6784	47.6871.
800	-5267.25	1.43979	27.5289
900	-5237.54	1.27259	18.7324
1000	-5292.44	1.15734	14.3662

$Cu_2CO_3 + SO_2 + 1/2\ O_2 \rightleftarrows Cu_2SO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-50814.2	37.2696	1.86047E+37
400	-41395.	22.6304	4.26997E+22
500	-32156.7	14.0639	1.15854E+14
600	-22952.7	8.36542	2.31961E+08
700	-13796.6	4.31003	20418.8
800	-4698.33	1.28428	19.2432
900	4333.87	-1.05302	8.85068E-02
1000	13292.3	-2.90673	1.23957E-03

TABLE XXV (Continued)

$Cu_2S + 2\ SO_2 \rightleftarrows Cu_2SO_4 + S_2$

```
T(K)          DEL F(CALS)    LOG10 K       K EQ
 298.15        9494.3        -6.96359       1.08745E-07
 400           21636.4       -11.8285       1.48423E-12
 500           33526.2       -14.6629       2.17320E-15
 600           45380.6       -16.5395       2.88704E-17
 700           57189.6       -17.8659       1.36189E-18
 800           68942.        -18.8451       1.42850E-19
 900           80626.1       -19.5902       2.56943E-20
 1000          92230.        -20.1687       6.78139E-21
```

$2\ Cu_2S + SO_2 \rightleftarrows 2\ Cu_2O + 3/2\ S_2$

```
T(K)          DEL F(CALS)    LOG10 K       K EQ
 298.15        80276.3       -58.8786       1.32241E-59
 400           81791.1       -44.7148       1.92847E-45
 500           83753.1       -36.6299       2.34474E-37
 600           86076.4       -31.3717       4.24911E-32
 700           88688.3       -27.706        1.96803E-28
 800           91535.3       -25.0209       9.52938E-26
 900           94576.        -22.9796       1.04798E-23
 1000          97776.9       -21.3817       4.15278E-22
```

$2\ CuCO_3 + SO_2 \rightleftarrows Cu_2SO_4 + 2\ CO_2$

```
T(K)          DEL F(CALS)    LOG10 K       K EQ
 298.15       -24171.2        17.7283       5.34972E+17
 400          -22417.3        12.2554       1.80056E+12
 500          -20670.8        9.0405        1.09775E+09
 600          -18957.9        6.90947       8.11846E+06
 700          -17308.7        5.40719       255381.
 800          -15743.4        4.30341       20109.9
 900          -14278.1        3.46922       2945.93
 1000         -12927.         2.82685       671.196
```

TABLE XXVI

THERMODYNAMIC VALUES FOR IRON COMPOUNDS

			Cp(T)			
Fe(+2)	H_f°	S_f°	A	B	C	D
FeO	-63.762	14.191	11.656	7.9993-3		7.597+4
$Fe(OH)_2$	-135.70	18.873				
$FeCO_3$	-178.55	22.194	11.625	2.678 -2		
$FeSO_3$	-165.54	25.3	18.57	17.1 -2		7.6 +4
$FeSO_4$	-220.41	25.682	20.96	1.974 -2		1.34 +5
FeS (138)	-22.8	15.200	12.2	2.38 -3		(ΔH=1.195)
	"	"				(ΔH=5.686)
Fe_2SiO_4	-343.54	35.38	36.51	9.36 -3		-6.70 +5
FeS_2	-42.45	12.70	18.0	1.182 -3		3.12 +5
Fe(+3)						
Fe_2O_3	-196.42	20.88	23.48	1.859 -2		3.548+5
$Fe_2(CO_2)_3$	-502.66	44.8	40.40	6.872 -2		7.72 +5
$Fe_2(SO_3)_3$	-457.4	53.8	44.21	6.389 -2		3.55 +5
$Fe_2(SO_4)_3$	-615.6	61.90	51.38	7.181 -2		5.290+5
$Fe(OH)_3$	-197.0					

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9 16 x 10^{-3}.

TABLE XXVII

REACTIONS OF IRON COMPOUNDS

$FeCO_3 + SO_2 \rightleftarrows FeSO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-8567.57	6.28388	1.92257E+06
400	-3783.91	2.06865	117.124
500	688.45	-.301098	.499922
600	4973.19	-1.81254	1.53977E-02
700	9088.28	-2.83915	1.44827E-03
800	13043.3	-3.56536	2.72047E-04
900	16843.	-4.09244	8.08283E-05
1000	20488.9	-4.48047	3.30771E-05

$1/2\ Fe_2SiO_4 + SO_2 \rightleftarrows FeSO_3 + 1/2\ SiO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-17653.3	12.9478	8.86715E+12
400	-6697.54	3.66151	4586.84
500	4156.74	-1.81798	1.52063E-02
600	15167.	-5.52782	2.96609E-06
700	26354.	-8.23291	5.84912E-09
800	37727.4	-10.3127	4.86758E-11
900	49293.6	-11.9771	1.05404E-12
1000	61058.4	-13.3521	4.44508E-14

$FeCO_3 + 1/2\ O_2 + SO_2 \rightleftarrows FeSO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-56245.7	41.2534	1.79233E+41
400	-53641.5	29.3255	2.11608E+29
500	-51312.1	22.4416	2.76467E+22
600	-49167.1	17.9196	8.31004E+17
700	-47185.5	14.7406	5.50333E+14
800	-45355.9	12.3979	2.49986E+12
900	-43672.	10.6112	4.08529E+10
1000	-42131.	9.21313	1.63354E+09

TABLE XXVII (Continued)

$1/2\ Fe_2SiO_4 + 1/2\ O_2 + SO_2 \rightleftarrows FeSO_4 + 1/2\ SiO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-65331.4	47.9173	8.26647E+47
400	-56555.1	30.9184	8.28703E+30
500	-47843.8	20.9248	8.40941E+20
600	-38973.3	14.2043	1.60078E+14
700	-29919.8	9.34687	2.22263E+09
800	-20671.8	5.65058	447285.
900	-11221.4	2.72652	532.741
1000	-1561.57	.341482	2.19524

$FeS + 2\ SO_2 \rightleftarrows FeSO_4 + S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-8894.56	6.52371	3.33975E+06
400	-3421.83	1.8707	74.2499
500	1790.34	-.783016	.16481
600	6872.51	-2.50478	3.12769E-03
700	11834.1	-3.69694	2.00935E-04
800	16676.4	-4.55844	2.76411E-05
900	21396.4	-5.1988	6.32710E-06
1000	25988.9	-5.68321	2.07393E-06

$2\ FeS + SO_2 \rightleftarrows 2\ FeO + 3/2\ S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	29185.8	-21.4063	3.92341E-22
400	27253.4	-14.8993	1.26101E-15
500	25191.6	-11.0177	9.60043E-12
600	23002.8	-8.38367	4.13360E-09
700	20702.8	-6.46749	3.40806E-07
800	18299.1	-5.00202	9.95362E-06
900	15794.9	-3.83776	1.45290E-04
1000	13190.5	-2.8844	1.30474E-03

TABLE XXVII (Continued)

$1/3\ Fe_2(CO_3)_3 + SO_2 \rightleftarrows 1/3\ Fe_2(SO_3)_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-6460.72	4.73861	54778.9
400	-6189.11	3.38355	2418.54
500	-5929.08	2.59312	391.848
600	-5706.57	2.07984	120.181
700	-5538.21	1.73012	53.7183
800	-5435.76	1.48585	30.6091
900	-5409.	1.31425	20.6184
1000	-5466.85	1.19548	15.6849

$1/3\ Fe_2(CO_3 + 1/2\ O_2 + SO_2 \rightleftarrows 1/3\ Fe_2(SO_4)_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-52688.	38.644	4.40536E+38
400	-50158.4	27.4214	2.63851E+27
500	-47684.6	20.8551	7.16378E+20
600	-45245.1	16.4902	3.09154E+16
700	-42853.5	13.3873	2.43954E+13
800	-40519.6	11.0759	1.19109E+11
900	-38251.9	9.29428	1.96914E+09
1000	-36057.9	7.88508	7.67499E+07

$Fe_2(CO_3)_3 + 2\ SO_2 \rightleftarrows FeSO_3 + FeSO_4 + 3\ CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-35714.1	26.1945	1.56497E+26
400	-36651.	20.0369	1.08873E+20
500	-37573.6	16.433	2.71031E+16
600	-38603.7	14.0696	1.17386E+14
700	-39790.6	12.4305	2.69451E+12
800	-41165.4	11.2525	1.78834E+11
900	-42751.5	10.3876	2.44104E+10
1000	-44569.7	9.74642	5.57723E+09

TABLE XXVII (Continued)

$$Fe_2(CO_3)_3 + 1/2\ O_2 + 2\ SO_2 \rightleftarrows 2\ FeSO_4 + 3\ CO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-83392.3	61.164	1.45895E+61
400	-86508.6	47.2938	1.96701E+47
500	-89574.1	39.1758	1.49885E+39
600	-92743.9	33.8018	6.33523E+33
700	-96064.4	30.0103	1.02390E+30
800	-99564.6	27.2157	1.64332E+27
900	-103267.	25.0912	1.23377E+25
1000	-107190.	23.44	2.75436E+23

$$Fe_2(SO_4)_3 + SO_2 + 2\ FeO \rightleftarrows 4\ FeSO_4$$

T(K)	DEL F(CALS)	LOG10 K	K.EQ
298.15	-53600.4	39.3132	2.05670E+39
400	-49563.9	27.0963	1.24827E+27
500	-45633.9	19.9583	9.08382E+19
600	-41750.7	15.2166	1.64657E+15
700	-37920.1	11.8461	7.01688E+11
800	-34146.4	9.33383	2.15689E+09
900	-30432.8	7.39442	2.47983E+07
1000	-26782.7	5.85678	719086.

$$FeS_2 + 2\ SO_2 \rightleftarrows 1.5\ S_2 + FeSO_4$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	17304.	-12.6916	2.03424E-13
400	19561.	-10.6939	2.02361E-11
500	21772.2	-9.52219	3.00476E-10
600	23943.2	-8.7264	1.87757E-09
700	26053.3	-8.13896	7.26169E-09
800	28087.	-7.67752	2.10125E-08
900	30031.3	-7.29688	5.04801E-08
1000	31874.	-6.97014	1.07118E-07

$$2\ FeS_2 + SO_2 \rightleftarrows 2\ FeO + 2.5\ S_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	81582.9	-59.837	1.45559E-60
400	73218.9	-40.0284	9.36654E-41
500	65155.3	-28.4961	3.19111E-29
600	57144.1	-20.8269	1.48961E-21
700	49141.1	-15.3515	4.45114E-16
800	41120.5	-11.2402	5.75207E-12
900	33064.8	-8.03393	9.24841E-09
1000	24960.6	-5.45834	3.48062E-06

TABLE XXVIII

THERMODYNAMIC VALUES FOR LITHIUM COMPOUNDS

			Cp(T)			
	H_f^o	S_f^o	A	B	C	D
Li_2O	-142.5	9.0519	14.932	6.0776-3		3.378+5
$LiOH_2$	-116.535	10.227	11.98	8.237 -3		2.267+5
Li_2CO_3	-290.26	21.582	20.57	2.279 -2		4.77 +5
$LiSO_3$	-279.4	26.00	21.84	2.118 -2		3.38 +5
$LiSO_4$	-342.58	35.357	24.23	2.382 -2		3.96 +5
$LiSiO_3$	-376.5	20.00				

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16 x 10^{-3}.

TABLE XXIX

REACTIONS OF LITHIUM COMPOUNDS

$$Li_2CO_3 + SO_2 \rightleftarrows Li_2SO_3 + CO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-11108.7	8.1477	1.40509E+08
400	-10981.6	6.00359	1.00829E+06
500	-10863.4	4.75118	56387.4
600	-10782.8	3.92992	8509.85
700	-10756.3	3.36022	2292.04
800	-10795.6	2.95096	893.229
900	-10910.7	2.65104	447.757
1000	-11110.4	2.42961	268.911

$$2\ LiOH + SO_2 \rightleftarrows Li_2SO_3 + H_2O$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-30603.2	22.4459	2.79190E+22
400	-29854.7	16.3214	2.09619E+16
500	-29062.6	12.7107	5.13670E+12
600	-28240.8	10.2927	1.96217E+10
700	-27406.6	8.56175	364546265
800	-26572.8	7.26361	1.83488E+07
900	-25749.7	6.25656	1.80533E+06
1000	-24946.5	5.45524	285262.

$$Li_2CO_3 + SO_2 + 1/2\ O_2 \rightleftarrows Li_2SO_4 + CO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-69772.8	51.1748	1.49569E+51
400	-68065.8	37.2112	1.62633E+37
500	-66399.5	29.0402	1.09703E+29
600	-64767.6	23.6054	4.03093E+23
700	-63183.6	19.7384	5.47474E+19
800	-61657.3	16.8539	7.14282E+16
900	-60197.2	14.6264	4.23093E+14
1000	-58810.8	12.8606	7.25480E+12

$$2\ LiOH + SO_2 + 1/2\ O_2 \rightleftarrows Li_2SO_4 + CO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-89267.3	65.473	2.97192E+65
400	-86938.9	47.5291	3.38107E+47
500	-84598.6	36.9997	9.99358E+36
600	-82225.6	29.9682	9.29439E+29
700	-79833.9	24.9399	8.70750E+24
800	-77434.5	21.1665	1.46729E+21
900	-75036.2	18.232	1.70589E+18
1000	-72646.8	15.8863	7.69593E+15

TABLE XXX

THERMODYNAMIC VALUES FOR MAGNESIUM COMPOUNDS

	H_f°	S_f°	Cp(T)			
			A	B	C	D
MgO	-143.63	6.4025	10.175	1.7392-3		1.4788+5
$Mg(OH)_2$	-220.82	15.077	13.034	1.579 -3		
$MgCO_3$	-263.98	15.696	18.613	1.3794-2		4.1592+5
$MgSO_3$	-241.0	22.5	17.09	1.684 -2		1.48 +5
$MgSO_4$	-305.5	21.9	16.53	2.18 -2		1.578 +3
MgS	-83.0	10.6	9.24	2.5 -3		7.93 +2
$MgSiO_3$	-357.63	16.197	24.535	5.437 -3		5.867 +5
$Mg_3(PO_4)_2$	-960.617					

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16 x 10^{-3}.

TABLE XXXI

REACTIONS OF MAGNESIUM COMPOUNDS

$MgCO_3 + SO_2 \rightarrow MgSO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	299.869	-.219939	.602644
400	259.126	-.141663	.721667
500	327.463	-.143218	.719088
600	436.001	-.158907	.693575
700	549.44	-.171644	.673529
800	644.685	-.176223	.666464
900	704.353	-.171141	.67431
1000	714.084	-.156154	.697984

$Mg(OH)_2 + SO_2 \rightarrow MgSO_3 + H_2O$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-5012.82	3.67665	4749.55
400	-4007.01	2.19061	155.1
500	-3209.1	1.40352	25.3233
600	-2549.87	.929333	8.49832
700	-2006.71	.626891	4.23537
800	-1568.11	.428639	2.68311
900	-1228.13	.298406	1.98795
1000	-984.035	.215187	1.6413

$MgSiO_3 + SO_2 \rightarrow MgSO_3 + SiO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-17104.3	12.5452	3.50889E+12
400	-10829.7	5.92054	832793.
500	-3992.22	1.74602	55.7215
600	3416.53	-1.2452	5.68592E-02
700	11347.4	-3.54489	2.85173E-04
800	19776.1	-5.40575	3.92867E-06
900	28693.	-6.9717	1.06734E-07
1000	38097.5	-8.33169	4.66567E-09

$MgCO_3 + SO_2 + 1/2\ O_2 \rightarrow MgSO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-56715.5	41.598	3.96255E+41
400	-54220.1	29.6418	4.38361E+29
500	-51520.2	22.5326	3.40916E+22
600	-48683.4	17.7433	5.53744E+17
700	-45766.6	14.2974	1.98318E+14
800	-42805.5	11.7008	5.02068E+11
900	-39825.2	9.67654	4.74837E+09
1000	-36845.5	8.0573	1.14103E+08

TABLE XXXI (Continued)

$$Mg(OH)_2 + SO_2 + 1/2\ O_2 \rightarrow MgSO_4 + H_2O$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-62028.2	45.4946	3.12296E+45
400	-58486.2	31.9741	9.42121E+31
500	-55056.7	24.0794	1.20056E+24
600	-51669.3	18.8315	6.78498E+18
700	-48322.8	15.0959	1.24709E+15
800	-45018.2	12.3056	2.02127E+12
900	-41757.7	10.1461	1.39988E+10
1000	-38543.6	8.42864	268311654

$$MgSiO_3 + SO_2 + 1/2\ O_2 \rightarrow MgSO_4 + SiO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-74119.7	54.3631	2.30719E+54
400	-65308.9	35.704	5.05862E+35
500	-55839.8	24.4219	2.64172E+24
600	-45702.9	16.657	4.53959E+16
700	-34968.7	10.9241	8.39678E+10
800	-23674.	6.47123	2.95958E+06
900	-11836.5	2.87599	751.601
1000	537.927	-.117633	.762724

$$MgS \quad 2\ SO_2 \rightleftarrows S_2 \quad MgSO_4$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-34028.5	24.9582	9.08139E+24
400	-28426.	15.5404	3.47032E+15
500	-23100.9	10.1033	1.26859E+10
600	-17901.8	6.52454	3.34608E+06
700	-12813.2	4.0028	10064.6
800	-7831.98	2.14085	138.309
900	-2960.71	.719381	5.2406
1000	1795.21	-.392573	.404974

$$2\ MgS + SO_2 \rightleftarrows MgO + 3/2\ S_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-8248.87	6.05013	1.12235E+06
400	-9477.11	5.18109	151736.
500	-11116.7	4.86194	72768.4
600	-13038.3	4.75197	56489.3
700	-15175.	4.74063	55033.9
800	-17491.9	4.78137	60446.2
900	-19969.2	4.85202	71124.7
1000	-22595.3	4.9411	87317.8

TABLE XXXII

THERMODYNAMIC VALUES OF MANGANESE COMPOUNDS

			Cp(T)			
	H_f°	S_f°	A	B	C	D
MnO	-91.93	14.265	11.104	1.94 -3		8.79 +4
$Mn(OH)_2$	-165.677	21.095				
$MnCO_3$	-213.74	20.474	21.99	9.3 -3		4.69 +5
$MnSO_3$	-197.37	24.40	18.01	1.704-2		8.8 +4
$MnSO_4$	-253.95	26.781	29.241	8.916-3		7.038+5
MnS	-49.452	18.682	11.396	1.799-3		
$MnSiO_3$	-302.21	21.2836	26.398	38.77 -3		6.156+5
Mn_2O_3	-229.11	26.398	24.726	8.376-3		3.228+5
$Mn(OH)_3$	-212.0					
Mn_2CO_3	-545.46	44.6	41.65	5.851-2		7.4 +5
$Mn_2(SO_3)_3$	-502.39	53.6	45.46	5.368-2		3.23 +5
$Mn_2(SO_4)_3$	-666.9	61.7	52.63	6.16 -2		4.97 +5

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16 x 10^{-3}.

TABLE XXXIII

REACTIONS OF MANGANESE COMPOUNDS

$MnCO_3 + SO_2 \rightleftarrows MnSO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-5452.06	3.99881	9972.58
400	-5115.56	2.79665	626.114
500	-4575.63	2.00118	100.272
600	-3927.09	1.43128	26.9949
700	-3221.7	1.00645	10.1496
800	-2492.51	.681322	4.80089
900	-1763.6	.428512	2.68233
1000	-1054.12	.230513	1.70025

$MnSiO_3 + SO_2 \rightleftarrows MnSO_3 + SiO_2$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-27944.3	20.4957	3.13113E+20
400	-22412.3	12.2527	1.78935E+12
500	-16249.9	7.10698	1.27931E+07
600	-9482.34	3.45596	2857.35
700	-2168.44	.677414	4.75789
800	5662.14	-1.54773	2.83314E-02
900	13996.2	-3.40074	3.97433E-04
1000	22830.9	-4.99261	1.01717E-05

$MnSiO_3 + 1/2\ O_2 + SO_2 \rightleftarrows MnSO_4 + SiO_2$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-77928.4	57.1566	1.43408E+57
400	-70095.3	38.3208	2.09299E+38
500	-62091.4	27.156	1.43226E+27
600	-53750.1	19.5899	3.88988E+19
700	-45057.4	14.0758	1.19069E+14
800	-35999.8	9.84045	6.92548E+09
900	-26562.	6.45393	2.84397E+06
1000	-16726.8	3.65778	4547.62

$MnCO_3 + 1/2\ O_2 + SO_2 \rightleftarrows MnSO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-55436.2	40.6597	4.56752E+40
400	-52798.6	28.8647	7.32364E+28
500	-50417.1	22.0502	1.12262E+22
600	-48194.8	17.5653	3.67497E+17
700	-46110.6	14.4048	2.53999E+14
800	-44154.4	12.0695	1.17356E+12
900	-42321.8	10.2832	1.91944E+10
1000	-40611.8	8.88091	760161134

TABLE XXXIII (Continued)

$MnS + 2\ SO_2 \rightleftarrows MnSO_4 + S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-15072.1	11.0546	1.13398E+11
400	-9323.66	5.0972	125083.
500	-4265.8	1.86567	73.396
600	384.166	-.140014	.724412
700	4707.18	-1.47051	3.38446E-02
800	8746.7	-2.39089	4.06548E-03
900	12527.5	-3.04387	9.03914E-04
1000	16063.8	-3.51281	3.07037E-04

$2\ MnS + SO_2 \rightleftarrows 2\ MnO + 3/2\ S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	28186.	-20.673	2.12312E-21
400	26627.5	-14.5571	2.77247E-15
500	24912.	-10.8954	1.27230E-11
600	23055.2	-8.40279	3.95558E-09
700	21076.6	-6.58428	2.60448E-07
800	18985.7	-5.18969	6.46113E-06
900	16786.9	-4.07881	8.34054E-05
1000	14481.8	-3.16684	6.81018E-04

$1/3\ Mn_2(CO_3)_3 + SO_2 \rightleftarrows 1/3\ Mn_2(SO_3)_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-7190.65	5.27398	187922.
400	-6919.03	3.7826	6061.79
500	-6659.01	2.91236	817.251
600	-6436.5	2.34587	221.752
700	-6268.14	1.95815	90.8133
800	-6165.69	1.68537	48.459
900	-6138.92	1.49161	31.0176
1000	-6196.78	1.3551	22.6516

$1/3\ Mn_2(CO_3) + 1/2\ O_2 + SO_2 \rightleftarrows 1/3\ Mn_2(SO_4)_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-55521.	40.7219	5.27085E+40
400	-52991.5	28.9702	9.33627E+28
500	-50517.6	22.0942	1.24221E+22
600	-48078.1	17.5227	3.33210E+17
700	-45686.5	14.2723	1.87216E+14
800	-43352.7	11.8504	7.08517E+11
900	-41084.9	9.98264	9.60811E+09
1000	-38891.	8.5046	319597559

TABLE XXXIII (Continued)

$Mn_2(CO_3)_3 + 2SO_2 \rightleftarrows Mn \quad MnSO_4 + 3\ CO_2$

```
T(K)              DEL F(CALS)     LOG10 K          K EQ
 298.15           -58403.1         42.8357          6.85039E+42
 400              -63651.9         34.7982          6.28287E+34
 500              -69181.1         30.2568          1.80618E+30
 600              -75050.4         27.3531          2.25494E+27
 700              -81243.5         25.3802          2.40014E+25
 800              -87753.6         23.9872          9.71020E+23
 900              -94580.6         22.9808          9.56687E+22
 1000             -101729.         22.2458          1.76130E+22
```

$Mn_2(CO_3)_3 + 2\ SO_2 + 1/2\ O_2 \rightleftarrows 2MnSO_4 + 3\ CO_2$

```
T(K)              DEL F(CALS)     LOG10 K          K EQ

OVERFLOW   IN 580
 298.15            -108387.         79.4966          5.78960E+76
 400               -111335.         60.8662          7.34906E+60
 500               -115023.         50.3058          2.02215E+50
 600               -119318.         43.4871          3.06978E+43
 700               -124132.         38.7786          6.00648E+38
 800               -129416.         35.3754          2.37362E+35
 900               -135139.         32.8354          6.84592E+32
 1000              -141286.         30.8962          7.87454E+30
```

$Mn_2(SO_4)_3 + 2MnO + SO_2 \rightleftarrows 4\ MnSO_4$

```
T(K)              DEL F(CALS)     LOG10 K          K EQ
 298.15           -81450.5          59.7399          5.49374E+59
 400              -77068.           42.1327          1.35732E+42
 500              -74415.1          32.5459          3.51459E+32
 600              -72854.1          26.5526          3.56969E+26
 700              -72115.9          22.5288          3.37898E+22
 800              -72043.3          19.6928          4.93003E+19
 900              -72534.9          17.6242          4.20936E+17
 1000             -73520.8          16.0774          1.19504E+16
```

TABLE XXXIV

THERMODYNAMIC VALUES OF MOLYBDENUM COMPOUNDS

			Cp(T)			
Mo(+4)	H_f°	S_f°	A	B	C	D
MoO_2	-140.83	11.0562				
MoS_2	-55.97	15.098				
Mo(+6)						
MoO_3	-177.885	18.572	20.06	5.898-3		3.679+5
MoS_3	-61.4689	15.887				

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16×10^{-3}.

TABLE XXXV

THERMODYNAMIC VALUES OF NICKEL COMPOUNDS

			Cp(T)			
Ni(+2)	H°_f	S°_f	A	B	C	D
NiO (250)	-57.264	9.078	4.988	3.7555-2		3.8893+5
(25-250)			11.176	2.0187-3		
$Ni(OH)_2$	-128.6	19.0				
$NiSO_3$	-155.86	25.4	27.69	7.28 -3		-5.8 +4
$NiSO_4$	-218.5	18.586	30.078	9.917 -3		
NiS	-17.48	16.10	9.245	12.795 -3		
Ni(+3)						
Ni_2S_3	-43.36	36.58				
$Ni(OH)_3$	-162.1					

Note: Positive and negative integers after values in Columns B an D are exponents of powers of 10. Thus, 9.16-3 is 9.16×10^{-3}.

TABLE XXXVI

REACTIONS OF NICKEL COMPOUNDS

$NiS + 2\ SO_2 \rightarrow S_2 + NiSO_4$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-9920.56	7.27623	1.88898E+07
400	-4551.94	2.48852	307.979
500	294.943	-.128995	.743027
600	4791.06	-1.74616	1.79405E-02
700	8977.22	-2.80446	1.56871E-03
800	12878.9	-3.52043	3.01699E-04
900	16512.4	-4.01211	9.72494E-05
1000	19887.7	-4.34901	4.47701E-05

$2\ NiS + SO_2 \rightarrow 2\ NiO + 3/2\ S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	35127.4	-25.7642	1.72122E-26
400	33216.6	-18.1593	6.92909E-19
500	31101.	-13.6022	2.49908E-14
600	28784.1	-10.4907	3.23049E-11
700	26287.5	-8.21214	6.13560E-09
800	23625.4	-6.45794	3.48387E-07
900	20806.6	-5.05551	8.80022E-06
1000	17836.9	-3.90055	1.25734E-04

TABLE XXXVII

THERMODYNAMIC VALUES FOR POTASSIUM COMPOUNDS

	H_f^o	S_f^o	A	B	C	D
K_2O	-86.362	23.484	17.91	9.32 -3		4.0 +4
KOH	-101.69					
K_2CO_3	-271.87	36.074	23.550	2.603 -2		1.79+5
K_2SO_3	-266.9	37.40	24.82	2.442 -2		4.0 +4
K_2SO_4	-342.34	41.975	28.76	2.379 -2		4.257+5
595°C ($\Delta H=2.25$)			33.59	1.3395-2		
K_2S	-87.867					

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16×10^{-3}.

TABLE XXXVIII

REACTIONS OF POTASSIUM COMPOUNDS

$$K_2CO_3 + SO_2 \rightarrow K_2SO_3 + CO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-16076.9	11.7916	6.18827E+11
400	-15634.8	8.54747	3.52751E+08
500	-15207.4	6.65106	4.47772E+06
600	-14817.6	5.40046	251454.
700	-14481.9	4.52409	33426.4
800	-14212.	3.88483	7670.56
900	-14017.9	3.40602	2546.92
1000	-13908.4	3.04147	1100.2

$$K_2CO_3 + SO_2 + 1/2\ O_2 \rightarrow K_2SO_4 + CO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-85575.2	62.7651	5.82211E+62
400	-82503.6	45.1043	1.27150E+45
500	-78986.9	34.5454	3.51078E+34
600	-75069.2	27.36	2.29070E+27
700	-70823.1	22.1249	1.33334E+22
800	-66305.5	18.1244	1.33181E+18
900	-61562.	14.9581	9.07949E+14
1000	-56630.5	12.3838	2.42012E+12

TABLE XXXIX

THERMODYNAMIC VALUES FOR SODIUM COMPOUNDS

			Cp(T)			
	H_f°	S_f°	A	B	C	D
Na_2O	102.87	16.986	15.693	5.3968-3		
$NaOH$	-101.91	27.71	-7.0231	5.5498-2	-2.036-5	5.8106+5
Na_2CO_3	-269.72	32.49	16.874	3.2395-2		
Na_2SO_3	-260.40	34.879	22.60	2.05 -2		
Na_2SO_4	-330.64	35.692	15.533	5.2773-2		
Na_2S	-92.96	22.48	19.81	1.64 -3		
Na_2SiO_3	-362.65	27.19	31.39	9.597 -3		6.467+5
Na_3PO_4	-288.35					

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16 x 10^{-3}.

TABLE XL

REACTIONS OF SODIUM COMPOUNDS

$Na_2CO_3 + SO_2 \rightleftarrows Na_2SO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-12043.8	8.83352	681583092
400	-11973.6	6.54589	3.51468E+06
500	-12072.1	5.27982	190468.
600	-12321.1	4.4906	30945.7
700	-12712.1	3.97123	9359.01
800	-13241.2	3.61944	4163.31
900	-13907.3	3.37913	2394.01
1000	-14711.5	3.21707	1648.44

$2\,NaOH + SO_2 \rightarrow Na_2SO_3 + H_2O$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-33075.4	24.2591	1.81596E+24
400	-34133.4	18.6606	4.57696E+18
500	-35915.2	15.7077	5.10160E+15
600	-38477.	14.0235	1.05550E+14
700	-41820.2	13.0645	1.16015E+13
800	-45942.2	12.5582	3.61561E+12
900	-50845.	12.3541	2.25992E+12
1000	-56536.7	12.3633	2.30846E+12

$Na_2CO_3 + 1/2\ O_2 + SO_2 \rightleftarrows Na_2SO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-75220.5	55.1704	1.48056E+55
400	-71701.4	39.1988	1.58041E+39
500	-68155.6	29.8083	6.43070E+29
600	-64557.	23.5287	3.37793E+23
700	-60930.2	19.0344	1.08252E+19
800	-57294.4	15.6613	4.58440E+15
900	-53665.2	13.0393	1.09480E+13
1000	-50056.4	10.9462	8.83539E+10

$Na_2SiO_3 + SO_2 \rightarrow Na_2SO_3 + SiO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-31897.6	23.3953	2.48463E+23
400	-25935.8	14.179	1.50997E+14
500	-19300.7	8.4413	2.76247E+08
600	-12032.7	4.38547	24292.1
700	-4200.14	1.31211	20.5169
800	4161.92	-1.13765	7.28366E-02
900	13037.2	-3.16773	6.79630E-04
1000	22420.9	-4.90296	1.25037E-05

TABLE XL (Continued)

$$2\ NaOH + SO_2 + 1/2\ O_2 \rightleftarrows NaSO_4 + H_2O$$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-96252.	70.596	3.94468E+70
400	-93861.2	51.3135	2.05807E+51
500	-91998.6	40.2361	1.72243E+40
600	-90712.9	33.0615	1.15215E+33
700	-90038.3	28.1277	1.34189E+28
800	-89995.4	24.6	3.98131E+24
900	-90603.	22.0143	1.03348E+22
1000	-91881.6	20.0925	1.23730E+20

$$Na_2SiO_3 + SO_2 + 1/2\ O_2 \rightleftarrows Na_2SO_3 + SiO_2$$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-95074.3	69.7322	5.39720E+69
400	-85663.6	46.8319	6.78971E+46
500	-75384.2	32.9697	9.32681E+32
600	-64268.5	23.4235	2.65165E+23
700	-52418.3	16.3753	2.37310E+16
800	-39891.3	10.9042	8.02034E+10
900	-26720.7	6.49248	3.10800E+06
1000	-12924.	2.82619	670.182

$$Na_2S + 2\ SO_2 \rightarrow Na_2SO_4 + S_2$$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	-49802.4	36.5275	3.36917E+36
400	-43255.3	23.6474	4.44060E+23
500	-36706.9	16.054	1.13233E+16
600	-30051.4	10.9526	8.96683E+10
700	-23312.7	7.28283	1.91793E+07
800	-16515.6	4.5145	32696.7
900	-9683.43	2.35284	225.339
1000	-2837.77	.620557	4.17404

$$2\ Na_2S + SO_2 \rightleftarrows 2\ Na_2O + 1.5\ S_2$$

T(K)	DEL F(CALS)	LOG10 K	K E0
298.15	93916.5	-68.883	1.30906E-69
400	93117.8	-50.9071	1.23865E-51
500	92257.7	-40.3495	4.47237E-41
600	91368.	-33.3003	5.00839E-34
700	90460.4	-28.2596	5.50079E-29
800	89535.	-24.4742	3.35609E-25
900	88587.5	-21.5246	2.98814E-22
1000	87611.8	-19.1588	6.93791E-20

TABLE XLI

THERMODYNAMIC VALUES FOR TIN COMPOUNDS

			Cp(T)			
Sn(+2)	H_f°	S_f°	A	B	C	D
SnO	-68.325	13.555	9.5464	3.4975-3		
$Sn(OH)_2$	-134.1	37.0				
SnS	-24.320	18.395	8.4795	7.428-3		9.526+4
Sn(+4)						
SnO_2	-138.75	12.504	17.652	2.3986-3		5.1579+5
SnS_2	-39.896	20.880	15.510	4.197 -3		9.63 +1
$Sn(CO_3)_2$	-330.04	33.9	28.94	3.58 -2		7.94 +5
$Sn(SO_3)_2$	-295.36	42.9	31.48	3.26 -2		5.16 +5
$Sn(SO_4)_2$	-393.40	37.1	36.26	3.788 -2		6.32 +5
$Sn(OH)_4$	-265.3					

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16×10^{-3}.

TABLE XLII

REACTIONS OF TIN COMPOUNDS

$$2\ SnS + SO_2 \rightleftarrows 2\ SnO + 3/2\ S_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	25384.3	-18.6181	2.40950E-19
400	23309.1	-12.743	1.80721E-13
500	21141.	-9.24612	5.67384E-10
600	18837.9	-6.86571	1.36234E-07
700	16402.3	-5.12403	7.51566E-06
800	13837.3	-3.78238	1.65051E-04
900	11144.8	-2.70792	1.95920E-03
1000	8325.98	-1.82071	.015111

$$1/2\ Sn(CO_3)_2 + SO_2 \rightleftarrows 1/2\ Sn(SO_3)_2 + CO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-4653.19	3.41288	2587.51
400	-4534.1	2.47877	301.141
500	-4423.82	1.93476	86.0565
600	-4351.06	1.5858	38.5304
700	-4332.45	1.35345	22.5656
800	-4379.75	1.19719	15.7468
900	-4502.74	1.09406	12.4181
1000	-4710.35	1.03005	10.7164

$$1/2\ Sn(CO_3)_2 + SO_2 + 1/2\ O_2 \rightleftarrows 1/2\ Sn(SO_4)_2 + CO_2$$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-45502.8	33.374	2.36613E+33
400	-42555.5	23.2649	1.84016E+23
500	-39671.4	17.3505	2.24142E+17
600	-36821.7	13.4202	2.63117E+13
700	-34019.9	10.6277	4.24334E+10
800	-31275.8	8.54916	3.54131E+08
900	-28597.9	6.94858	8.88342E+06
1000	-25993.7	5.68425	483337.

TABLE XLII (Continued)

$1/2\ SnS + 2\ SO_2 \rightleftharpoons S_2 + 1/2\ Sn(SO_4)_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	12670.6	-9.29328	5.09001E-10
400	18499.3	-10.1135	7.70095E-11
500	23958.	-10.4782	3.32525E-11
600	29223.9	-10.651	2.23338E-11
700	34324.9	-10.723	1.89240E-11
800	39272.5	-10.735	1.84062E-11
900	44070.4	-10.708	1.95872E-11
1000	48717.8	-10.6535	2.22067E-11

$1/2\ SnS_2 + SO_2 \rightleftharpoons SnO_2 + 1.5\ S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	14151.5	-10.3794	4.17456E-11
400	12730.6	-6.95977	1.09705E-07
500	10965.6	-4.79586	1.60008E-05
600	8947.75	-3.26113	5.48118E-04
700	6730.05	-2.10245	7.89865E-03
800	4340.71	-1.18652	6.50644E-02
900	1795.87	-.436352	.36614
1000	-895.113	.195742	1.56943

TABLE XLIII

THERMODYNAMIC VALUES FOR ZINC COMPOUNDS

			Cp(T)			
	H°_f	S°_f	A	B	C	D
ZnO	-83.376	10.392	11.704	1.2184-3		2.1788+5
$Zn(OH)_2$	-153.37					
$ZnCO_3$	-193.68	19.685	9.2932	3.2968-2		
$ZnSO_3$	-178.48	25.8	18.610	2.728 -2		2.18 +5
$ZnSO_4$	-233.69	29.767	17.062	2.0791-2		
ZnS	-48.5	13.800	12.16	1.236 -3		1.361 +5
$ZnSiO_3$	-294.32	20.498				

Note: Positive and negative integers after values in Columns B and D are exponents of powers of 10. Thus, 9.16-3 is 9.16 x 10^{-3}.

TABLE XLIV

REACTIONS OF ZINC COMPOUNDS

$ZnCO_3 + SO_2 \rightleftarrows ZnSO_3 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-7274.71	5.33563	216586.
400	-7265.67	3.9721	9377.84
500	-7589.86	3.31947	2086.74
600	-8173.1	2.97879	952.343
700	-8979.22	2.80508	638.386
800	-9987.85	2.73015	537.223
900	-11187.2	2.71821	522.655
1000	-12570.7	2.74893	560.954

$ZnCO_3 + SO_2 + 1/2\ O_2 \rightleftarrows ZnSO_4 + CO_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	-56361.7	41.3385	2.18022E+41
400	-54674.9	29.8905	7.77161E+29
500	-53160.9	23.2503	1.77931E+23
600	-51777.5	18.871	7.43015E+18
700	-50518.5	15.7818	6.05104E+15
800	-49381.	13.4982	3.14891E+13
900	-48364.2	11.7513	5.64062E+11
1000	-47469.3	10.3805	2.40155E+10

$ZnS + 2\ SO_2 \rightleftarrows ZnSO_4 + S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	1890.08	-1.38628	4.10884E-02
400	6700.72	-3.66325	2.17146E-04
500	11452.8	-5.00895	9.79605E-06
600	16197.6	-5.90342	1.24905E-06
700	20912.1	-6.53289	2.93166E-07
800	25578.7	-6.99187	1.01889E-07
900	30182.1	-7.33351	4.63973E-08
1000	34708.5	-7.58998	2.57051E-08

$2\ ZnS + SO_2 \rightleftarrows 2\ ZnO + 1.5\ S_2$

T(K)	DEL F(CALS)	LOG10 K	K EQ
298.15	42788.4	-31.3831	4.13903E-32
400	41069.4	-22.4196	3.80517E-23
500	39089.9	-17.0962	8.01294E-18
600	37037.9	-13.4989	3.16998E-14
700	34870.6	-10.8935	1.27797E-11
800	32596.5	-8.91016	1.22981E-09
900	30219.1	-7.34249	4.54473E-08
1000	27739.3	-6.06597	8.59067E-07

APPENDIX III

COMPUTER PROGRAMS

This appendix contains some of the computer programs used in this survey.

Computer Program I is the program which lists the total file of reference cards in order alphabetically by author and numbers them sequentially. The last page of this program contains the instructions for making the print-out, as described in the text of the report.

Computer Program II is the program used to search for certain reference numbers and to arrange these in alphabetical order, according to any one of the other categories. Similar programs have been written to search for references by author, date, journal, or type of reference, as described above.

Computer Programs III and IV were used to calculate the thermodynamics of sulfur dioxide sorption as described in Section IV of the text.

COMPUTER PROGRAM I

```
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C   THIS PROGRAM LISTS THE FILE OF CARDS IN ORDER ALPHABETICALLY BY AUTHOR,
C   DESCRIBED IN III IN THE GENERAL FORMAT (FORMAT 1).  IT NUMBERS THEM
C   SEQUENTIALLY.
      DIMENSION INAMF(10),ISH(20)
      COMMON IESC(75),I1(75),I2(75),I3(75),I4(75),I5(75),I6(75)
      CALL REREAD
      KK=0
      LINE=0
      READ(1,1)IPAR,INAME
    1 FORMAT(A3,10A3)
      READ(1,999)LAST,LDASH,KWIC,LCHECK,LBLANK
  999 FORMAT(5A1)
      WRITE(3,2)IPAR,INAME
    2 FORMAT(1H1,25X,A3,3X,10A3,/)
      J=0
C***********************************************************************
    4 READ(1,3)IESC,IREF,NUM
    3 FORMAT(75A1,A4,I1)
      IF(NUM-9)10,11,10
   10 IF(NUM)456,455,456
  455 STOP
  456 J=J+1
      CALL CHANGE(LAST,LDASH,KWIC,LCHECK,LBLANK,J)
      GO TO 4
   11 READ(99,5)IK,ISH,IREF,NUM
      KK=KK+1
    5 FORMAT(A2,20A3,13X,A4,I1)
  802 GO TO (300,301,302,303,304,305),J
  300 IF(LINE-42)700,700,701
  701 WRITE(3,555)
  555 FORMAT(1H1,1X)
      LINE=0
  700 LINE=LINE+1+J
      IF(I1(1)-LBLANK)50,51,50
   50 WRITE(3,522)KK,I1
      WRITE(3,8)IK,ISH,IREF
      GO TO 99
   51 WRITE(3,523)KK,I1
      WRITE(3,8)IK,ISH,IREF
      GO TO 99
  301 IF(LINE-42)702,702,703
  703 WRITE(3,555)
      LINE=0
  702 LINE=LINE+1+J
      IF(I1(1)-LBLANK)60,61,60
   60 WRITE(3,522)KK,I1
 6777 IF(I2(1)-LBLANK)62,63,62
   62 WRITE(3,9)I2
      WRITE(3,8)IK,ISH,IREF
      GO TO 99
   61 WRITE(3,523)KK,I1
      GO TO 6777
   63 WRITE(3,860)I2
      WRITE(3,8)IK,ISH,IREF
```

Computer Program I (Continued)

```
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      GO TO 99
  302 IF(LINE-42)704,704,705
  705 WRITE(3,555)
      LINE=0
  704 LINE=LINE+1+J
      IF(I1(1)-LBLANK)64,65,64
   64 WRITE(3,522)KK,I1
 7777 IF(I2(1)-LBLANK)66,67,66
   66 WRITE(3,9)I2
 8777 IF(I3(1)-LBLANK)68,71,68
   68 WRITE(3,9)I3
      WRITE(3,8)IK,ISH,IREF
      GO TO 99
   65 WRITE(3,523)KK,I1
      GO TO 7777
   67 WRITE(3,860)I2
      GO TO 8777
   71 WRITE(3,860)I3
      WRITE(3,8)IK,ISH,IREF
      GO TO 99
  303 IF(LINE-42)706,706,707
  707 WRITE(3,555)
      LINE=0
  706 LINE=LINE+1+J
      IF(I1(1)-LBLANK)72,73,72
   72 WRITE(3,522)KK,I1
 7888 IF(I2(1)-LBLANK)74,75,74
   74 WRITE(3,9)I2
 8888 IF(I3(1)-LBLANK)76,77,76
   76 WRITE(3,9)I3
 9888 IF(I4(1)-LBLANK)78,81,78
   78 WRITE(3,9)I4
 9000 WRITE(3,8)IK,ISH,IREF
      GO TO 99
   73 WRITE(3,523)KK,I1
      GO TO 7888
   75 WRITE(3,860)I2
      GO TO 8888
   77 WRITE(3,860)I3
      GO TO 9888
   81 WRITE(3,860)I4
      GO TO 9000
  304 IF(LINE-42)708,708,709
  709 WRITE(3,555)
      LINE=0
  708 LINE=LINE+1+J
      IF(I1(1)-LBLANK)82,83,82
   82 WRITE(3,522)KK,I1
 2222 IF(I2(1)-LBLANK)84,85,84
   84 WRITE(3,9)I2
 3222 IF(I3(1)-LBLANK)86,87,86
   86 WRITE(3,9)I3
 4222 IF(I4(1)-LBLANK)88,89,88
   88 WRITE(3,9)I4
```

Computer Program I (Continued)

```
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 5222 IF(I5(1)-LBLANK)90,91,90
   90 WRITE(3,9)I5
 9001 WRITE(3,8)IK,ISH,IREF
      GO TO 99
   83 WRITE(3,523)KK,I1
      GO TO 2222
   85 WRITE(3,860)I2
      GO TO 3222
   87 WRITE(3,860)I3
      GO TO 4222
   89 WRITE(3,860)I4
      GO TO 5222
   91 WRITE(3,860)I5
      GO TO 9001
  305 IF(LINE-42)710,710,711
  711 WRITE(3,555)
      LINE=0
  710 LINE=LINE+1+J
      IF(I1(1)-LBLANK)100,101,100
  100 WRITE(3,522)KK,I1
 6222 IF(I2(1)-LBLANK)102,103,102
  102 WRITE(3,9)I2
 7222 IF(I3(1)-LBLANK)104,105,104
  104 WRITE(3,9)I3
 8222 IF(I4(1)-LBLANK)106,107,106
  106 WRITE(3,9)I4
 9222 IF(I5(1)-LBLANK)108,109,108
  108 WRITE(3,9)I5
 9223 IF(I6(1)-LBLANK)110,111,110
  110 WRITE(3,9)I6
 9002 WRITE(3,8)IK,ISH,IREF
      GO TO 99
  101 WRITE(3,523)KK,I1
      GO TO 6222
  103 WRITE(3,860)I2
      GO TO 7222
  105 WRITE(3,860)I3
      GO TO 8222
  107 WRITE(3,860)I4
      GO TO 9222
  109 WRITE(3,860)I5
      GO TO 9223
  111 WRITE(3,860)I6
      GO TO 9002
  522 FORMAT(1H ,I3,2X,75A1)
    9 FORMAT(1H ,5X,75A1)
    8 FORMAT(1H ,5X,A2,(20(1X,A3)),1X,A4,/)
  523 FORMAT(1H ,I3,1X,75A1)
  860 FORMAT(1H ,4X,75A1)
   99 DO 69 N=1,75
      I1(N)=LBLANK
      I2(N)=LBLANK
      I3(N)=LBLANK
      I4(N)=LBLANK
```

Computer Program I (Continued)

```
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            I5(N)=LBLANK
            I6(N)=LBLANK
   69 CONTINUE
      I=20
      J=0
  801 L=I
  800 CONTINUE
      J=0
      I=0
      IF(L-20)4,888,4
  888 DO 79 N=1,75
      I1(N)=LBLANK
      I2(N)=LBLANK
      I3(N)=LBLANK
      I4(N)=LBLANK
      I5(N)=LBLANK
      I6(N)=LBLANK
   79 CONTINUE
      GO TO 4
      END
```

Computer Program I (Continued)

```
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      SUBROUTINE CHANGE(LAST,LDASH,KWIC,LCHECK,LBLANK,J)
      COMMON IESC(75),I1(75),I2(75),I3(75),I4(75),I5(75),I6(75)
      DO 100 I=1,75
      IF(IESC(I)-LAST)10,11,10
   10 IF(IESC(I)-LDASH)20,11,20
   20 IF(IESC(I)-KWIC)30,11,30
   30 IF(IESC(I)-LCHECK)40,11,40
   40 GO TO 100
   11 IESC(I)=LBLANK
  100 CONTINUE
      GO TO (61,62,63,64,65,66),J
   61 DO 661 I=1,75
      I1(I)=IESC(I)
  661 CONTINUE
      GO TO 70
   62 DO 662 I=1,75
      I2(I)=IESC(I)
  662 CONTINUE
      GO TO 70
   63 DO 663 I=1,75
      I3(I)=IESC(I)
  663 CONTINUE
      GO TO 70
   64 DO 664 I=1,75
      I4(I)=IESC(I)
  664 CONTINUE
      GO TO 70
   65 DO 665 I=1,75
      I5(I)=IESC(I)
  665 CONTINUE
      GO TO 70
   66 DO 666 I=1,75
      I6(I)=IESC(I)
  666 CONTINUE
   70 RETURN
      END
```

Computer Program II

```
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C     SAMPLE I.D. SEARCH PROGRAM - 1
C  GIVEN CERTAIN REFERENCE NUMBERS THIS PROGRAM WILL LIST THEEM IN
C  ALPHABETICAL ORDER.
      DIMENSION INUMS(20),KBS(75),M(75)
      WRITE(3,456)
  456 FORMAT(1H1,1X)
      READ(1,3)NUMBER
    3 FORMAT(I2)
      READ(1,1)INUMS
    1 FORMAT(20A4)
      READ(1,4)LAST,LDASH,KWIC,LCHECK,LBLANK
    4 FORMAT(5A1)
      N=0
    5 READ(1,2)KBS,IREF,JCARD
    2 FORMAT(75A1,A4,I1)
      IF(JCARD)8,8,68
   68 DO 99 I=1,NUMBER
      IF(INUMS(I)-IREF)80,81,80
   80 GO TO 99
   81 IF(JCARD-1)20,23,21
   23 N=N+1
   21 DO 98 J=1,75
      IF(KBS(J)-LAST)10,11,10
   10 IF(KBS(J)-LDASH)70,71,70
   70 IF(KBS(J)-KWIC)30,31,30
   30 IF(KBS(J)-LCHECK)40,41,40
   40 M(J)=KBS(J)
      GO TO 98
   11 M(J)=LBLANK
      GO TO 98
   71 M(J)=LBLANK
      GO TO 98
   31 M(J)=LBLANK
      GO TO 98
   41 M(J)=LBLANK
   98 CONTINUE
      IF(M(1)-LBLANK)60,61,60
   60 WRITE(3,12)M
   12 FORMAT(1H ,1X,75A1)
      GO TO 69
   61 WRITE(3,13)M
   13 FORMAT(1H ,75A1)
      GO TO 69
   99 CONTINUE
      IF(NUMBER-N)8,8,5
    8 STOP001
    6 READ(1,2)KBS,IREF,JCARD
      IF(JCARD-9)108,108,1000
  108 WRITE(3,54)KBS,IREF
      GO TO 5
  100 IF(JCARD)8,8,21
   54 FORMAT(1H ,1X,2A1,1X,3A1,1X,3A1,1X,3A1,1X,3A1,1X,3A1,1X,3A1,1X,
     13A1,1X,3A1,1X,49A1,2X,A4,/)
   20 WRITE(3,7)
```

Computer Program II (Continued)

```
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      7 FORMAT(1H1,'FINISH ABSTRACT')
        STOP003
     69 GO TO 6
        END
```

COMPUTER PROGRAM III

```
CPARG


100 FOR I=1TO4
200 READ T(I)
300 LET A(I)=T(I)↑2
400 LET B(I) = 1./T(I)↑2
500 PRINT T(I),A(I),B(I)
600 NEXT I
700 DATA 300,400,500,600,
800 END
```

```
SIMEQ4


100 DIM A(4,4),X(4,1),C(4,1) ,B(4,4)
200 MAT READ A,C
300 MAT PRINT A,C
400 MAT B=INV(A)
500 MAT X=B*C
600 MAT PRINT X
700 DATA 1,300,90000,1.11111 E-5
701 DATA 1,500,250000,4 E-6
702 DATA 1,700,490000,2.04082 E-6
703 DATA 1,900,810000,1.23457 E-6
704 DATA 7.0237,7.4315,7.8837,8.2129
800 END
```

```
CPCOMP


100 READ N,A,B,C,D
200 FOR K=1 TO N
300 READ T(K)
400 LET C(K) = A + B*T(K) + C*T(K)↑2 + D*T(K)↑-2
500 PRINT T(K),C(K)
501 NEXT K
550 DATA 1,1,1,1,1
551 DATA 2
600 END
```

COMPUTER PROGRAM IV

```
THERCO


100 DIM T(8)
110 DIM C(18)
120 DIM D(18)
130 DIM G(18,8)
140 DIM V(18,13)
150 DIM H(18)
160 DIM S(18)
170 DIM A(18)
180 DIM B(18)
190 READ M,N,R,T0,Z
200 FOR I=1 TO M
210 READ H(I),S(I),A(I),B(I),C(I),D(I)
220 NEXT I
230 FOR K = 1 TO N
240 READ T(K)
250 NEXT K
260 PRINT "THE VALUE OF H0, S0,A,B,C,D FOR THE CP ARE "
270 FOR I = 1 TO M
280 REM PRINT OUT STARTING VALUES
290 PRINT H(I);S(I);A(I);B(I);C(I);D(I)
300 NEXT I
310 FOR K =1 TO N
320 FOR I = 1 TO M
330 LET Q = H(I) + A(I)*(T(K)-T0)+B(I)*(T(K)↑2-T0↑2)
340 LET Q=Q+C(I)*(T(K)↑3-T0↑3)/3-D(I)*((1/T(K))-(1/T0))
350 LET E = S(I)+A(I)*LOG(T(K)/T0)+B(I)*(T(K)-T0)
360 LET E = E+C(I)*(T(K)↑2-T0↑2)/2-D*((1/T(K)↑2)-(1/T0↑2))/2
370 LET G(I,K)=Q-T(K)*E
380 NEXT I
390 NEXT K
400  FOR J=1 TO Z
410 FOR I = 1 TO M
420 READ V(I,J)
430 NEXT I
440 PRINT
450 PRINT
460 PRINT "STOIC. COEFF."
470 FOR I=1 TO M
480 PRINT V(I,J)
490 NEXT I
500 PRINT
510 PRINT "T(K)","DEL F(CALS)", "LOG10 K", "K EQ"
520 FOR K=1 TO N
530 LET F(K) = 0
540 FOR I = 1 TO M
550 LET F(K) = F(K) + G(I,K)*V(I,J)
560 NEXT I
570 LET P(K)= -F(K)/(R*T(K)*LOG(10))
580 LET W(K) = EXP(-F(K)/(R*T(K)))
590 PRINT T(K),F(K),P(K),W(K)
```

COMPUTER PROGRAM IV (Cont'd)

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```
THERCO CONTINUED


600 NEXT K
610 NEXT J
620 REM CONTROL PARAMETERS M, N, R , T0 ,E
630 DATA 17
640 DATA 8
650 DATA 1.986
660 DATA 298.15
670 DATA 9
680 REM HERE ARE THE DATA FOR THE COMMON SPECIES
690 REM H0, S0, A, B(1), C(T2), C(T-2)
700 REM ORDER  O2 , SO2 , H2O , SO3 , SIO2 , S2 , CO2
710 DATA 0 , 49.007, 5.179 , 5.24E-3 , 2.141E-6 , 41860
720 DATA -70947 , 59.297 , 7.0756 , 1.076E-2 , -4.848E-6,-29107
730 DATA -57798 , 45.105 , 6.954 , 2.569E-3 , 3.152E-7 , 24706
740 DATA -94470 , 61.345 , 9.054 , 1.679E-2 , -7.659E-6 , -113429
750 DATA -217500, 10. , -5.364 , 5.413E-2 , -3.396E-5,257565
760 DATA 30840 , 54.51 , 7.866,1.892E-3 , 8.996E-7 , -29053
770 DATA -94035 , 51.125 , 6.94 , 9.742E-3 , 3.667E-6 , -57464
790 REM HERE ARE THE DATA ON COPPER COMPOUNDS
800 REM CUO , CUCO3 , CUSO3 , CUSO4 , CUS
810 REM CU20 , CU2CO3 , CU2SO3 , CU2SO4 , CU2S
820DATA  -39490 , 10.187 , 9.267 , 4.797E-3 , 0 , 0
830 DATA -142100 , 21.023 , 14.91 , 2.151E-2 , 0 , 139000
840 DATA -127280 , 25.70 , 16.18 , 1.99E-2 , 0 , 0
850 DATA -184220 , 27.067 , 18.805 , 1.718E-2 , 0 , 2792
860 DATA -11600 , 15.9 , 10.6 , 2.64E-3 , 0 , 0
870 REM CUPROUS
880 DATA -40780 , 22.521 , 14.893 , 5.698E-3 , 0 , 0
890 DATA-142290 , 37.63 , 20.53 , 2.241E-2 , 0 , 1.39E+5
900 DATA -127060 , 40.60 , 21.8 , 2.08E-2 , 0 , 0
910 DATA -179510 , 38.463 , 24.19 , 23.4E-2 , 0 , 58000
920 DATA -19.931 , 28.883 , 19.492 , 0 , 0 , 0
930 REM CU2S DATA (900) APPLY ONLY TO THE 25 TO 103 C RANGE
940 REM HERE ARE THE TEMPERATURES
950 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
960REM STOICHIOMETRIC COEFFICIENTS
970 REM STANDARD SEQUENCE FOR CUPRIC (1,1,2)
980 DATA 0, -1, 0, 0, 0, 0, 1,   0, -1, 1, 0, 0,   0, 0, 0, 0, 0
990 DATA -.5, -1, 0, 0, 0, 0, 1,   0, -1, 0 ,1, 0,   0, 0, 0, 0, 0
1000 DATA 0, -2, 0, 0, 0, 1, 0,   0, 0, 0, 1, -1,    0, 0, 0, 0, 0
1010 DATA 0, -1, 0, 0, 0, 1.5, 0,   1, 0, 0, 0, -2,   0, 0, 0, 0, 0
1020 REM STANDARD SEQUENCE FOR CUPROUS (1,1,2)
1030 DATA 0, -1, 0, 0, 0, 0, 1,   0, 0, 0, 0, 0,   0, -1, 1, 0, 0
1040 DATA -.5, -1, 0, 0, 0, 0, 1,  0, 0, 0, 0, 0,  0, -1, 0, 1, 0
1050DATA 0, -2, 0, 0, 0, 1, 0,  0, 0, 0, 0, 0,  0, 0, 0, 1, -1
1060 DATA 0, -1, 0, 0, 0, 1.5, 0,  0, 0, 0, 0, 0, 1, 0, 0, 0, -2
1070 REM REDOX INVOLVING COPPER
1080 DATA 0, -1, 0, 0, 0, 0, 2, 0, -2, 0, 0, 0, 0, 0, 0, 1, 0
1090 END
```

COMPUTER PROGRAM IV (Cont'd)

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REGDAT

```
520 REM H0,S0,A,B,C,D, OF EACH COMPOUND
530 REM CPDS.   H2SO4  SO3  H2O  O2  C  SO2  CO2  CO  H2
540 DATA -194548 , 37.501 , 36.77 , 9.16E-3 , -2.088E-6 , -543200
550 DATA -94470 , 61.345 , 9.054 , 1.679E-2 , -7.659E-6 , -113429
560 DATA -57798 , 45.105 , 6.954 , 2.569E-3 , 3.152E-7 , 24706
570 DATA 0 , 49.007 , 5.179 , 5.24E-3 , -2.141E-6 , 41860
580 DATA 0 , 1.372 , -.221 , 9.963E-3 , -4.611E-6 , -27007
590 DATA -70947 , 59.297 , 7.0756 , 1.076E-2 , -4.848E-6,-29107
600 DATA -94035 , 51.125 , 6.94 , 9.742E-3 , -3.667E-6 , -57464
610 DATA -26416 , 41.218 , 5.735 , 2.703E-3 , -5.356E-7 , 42111
620 DATA 0 , 31.208 , 7.562 , -1.5E-3 , 1.193E-6 , -29332
630 REM READ IN TEMPERATURES
640 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
650 REM READ IN STOICHIOMETRIC COEFFICIENTS
660 DATA -1 , 1 , 1 , 0 , 0 , 0 , 0 , 0 , 0
670 DATA 0 , -1 , 0 , .5 , 0 , 1 , 0 , 0 , 0
680 DATA -2 , 0 , 2 , 0 , -1 , 2 , 1 , 0 , 0
690 DATA -1 , 0 , 1 , 0 , -1 , 1 , 0 , 1 , 0
700 DATA -1  , 0 , 1 , 0 , 0 , 1 , 1 , 0 , -1
710 DATA -1 , 0 , 2 , 0 , 0 , 1 , 1 , 0 , -1
720 DATA 0 , -1 , 0 , 0 , -1 , 1 , 0 , 1 , 0
730 DATA 0 , -1 , 0 , 0 , 0 , 1 , 1 , -1 , 0
740 DATA 0 , -1 , 1 , 0 , 0 , 1 , 0 , 0 , -1
750 DATA 0 , -2 , 0 , 0 , -1 , 2 , 1 , 0 , 0
800 END
```

BASDAT

```
680 REM HERE ARE THE DATA FOR THE COMMON SPECIES
690 REM H0, S0, A, B(T), C(T2), C(T-2)
700 REM ORDER  O2 , CO2 , H2O , SO3 , S102 , S2 , CO2
710 DATA 0 , 49.007, 5.179 , 5.24E-3 , 2.141E-6 , 41860
720 DATA -70947 , 59.297 , 7.0756 , 1.076E-2 , -4.848E-6,-29107
730 DATA -57798 , 45.105 , 6.954 , 2.569E-3 , 3.152E-7 , 24706
740 DATA -94470 , 61.345 , 9.054 , 1.679E-2 , -7.659E-6 , -113429
750 DATA -217500, 10. , -5.364 , 5.413E-2 , -3.396E-5,257565
760 DATA 30840 , 54.51 , 7.866,1.892E-3 , 8.996E-7 , -29053
770 DATA -94035 , 51.125 , 6.94 , 9.742E-3 , 3.667E-6 , -57464
```

COMPUTER PROGRAM IV (Cont'd)

- 4 -

LI/DAT

```
790 REM HERE ARE THE DATA ON LITHIUM COMPOUNDS
800 REM LI20 , LIOH , LI2CO3 , LI2SO3 , LI2SO4
810 DATA -142500 , 9.0519 , 14.932 , 6.0776E-3 , 0 , 3.378E+5
820 DATA -116535 , 10.227 , 11.98 , 8.237E-3 , 0 , 2.267E+5
830 DATA -290260 , 21.582 , 20.57 , 2.279E-2 , 0 , 4.77E+5
840 DATA -279400 , 26.00 , 21.84 , 2.118E-2 , 0 , 3.38E+5
850 DATA -342580 , 35.357 , 24.23 , 2.382E-2 , 0 , 3.96E+5
910 REM HERE ARE THE STOICHIOMETRIC COEFFICIENTS
920 REM ACID REPLACEMENT REACTIONS
930 DATA 0 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , 0 , -1 , 1 , 0
940 DATA 0 , -1 , 1 , 0 , 0 , 0 , 0 , 0 , -2 , 0 , 1 , 0
950 REM NO SILICATE DATA
960 REM OXIDATIVE REPLACEMENT REACTIONS
970 DATA -.5 , -1 , 0 , 0 , 0 , 0,1 , 0 , 0 , -1 , 0 , 1
980 DATA -.5 , -1 , 1 , 0 , 0 , 0 , 0  ,0 , -2 , 0 , 0 , 1
990 REM NO SILICATE DATA
1000 REM REDOX REACTIONS ON SULFUR
1010 REM NO SULFIDE DATA
1020 REM NO SULFIDE DATA
```

NA/DAT

```
790 REM HERE ARE THE DMTA FOR SODIUM COMPOUNDS
800 REM NA2O , NAOH , OA2CO3 , NA2SO3 , NA2SO4 , NA2S , NA2SIO3
810 REM SULFATE HAS A PHASE TRANS AT 259C, H=2640
820 DATA -102870 , 16.986 , 15.693 , 5.3968E-3 , 0 , 0
830 DATA -101910 , 27.71 , -7.0231 , 5.54976E-2 , -2.036E-5 , 581057
840 DATA -269720 , 32.49 , 16.874 , 3.2395E-2 , 0 , 0
850 DATA -260400 , 34.879 , 22.60 , 2.05E-2 , 0 , 0
860 DATA -330640 , 35.692 , 15.533 , 5.2773E-2 , 0 , 0
870 DATA -92960 , 22.40 , 19.81 , 1.64E-3 , 0 , 0
880 DATA -362650 , 27.19 , 31.39 , 9.597E-3 , 0 , 6.467E+5
890 REM
900 REM READ IN TEMPERATURES
910 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
920 REM HERE ARE THE STOICHIOMETRIC COEFFICIENTS
930 REM ACID REPLACEMENT REACTIONS
940 DATA 0 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , 0 , -1 , 1 , 0 , 0 , 0
950 DATA 0,  -1 , 1 , 0 , 0 , 0 , 0 , 0 , -2 , 0 , 1 , 0 , 0 , 0
960 DATA 0 , -1 , 0 , 0 , 1 , 0 , 0 , 0 , 0 , 0 , 1 , 0 , 0 , -1
970 REM OXIDATIVE REPLACEMENT REACTIONS
980 DATA -.5 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , 0 , -1 , 0,1,0 ,0
990 DATA -.5 , -1 , 1 , 0 , 0 , 0 , 0 , 0 , -1 , 0 , 0 , 1 , 0 , 0
```

COMPUTER PROGRAM IV (Cont'd)

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```
NA/DAT CONTINUED


1000 DATA -.5 , -1 , 0 , 0 , 1 , 0 , 0 , 0 , 0 , 0 , 0 , 1 , 0 , -1
1010 REM REDOX REACTIONS ON SULFUR
1020 DATA 0 , -2 , 0 , 0 , 0 , 1 , 0 , 0 , 0 , 0 , 0 , 1 , -1 , 0 ,
1030 DATA 0 , -1 , 0 , 0 , 0 , 1.5 , 0 , 2 , 0 , 0 , 0 , 0 , -2 , 0
1040 END
```

```
K/DAT


790 REM HERE ARE THE DATA ON POTASSIUM COMPOUNDS
800 REM K20 , K2CO3 , K2SO3 , K2SO4 ,
810 DATA -86362 , 23.484 , 17.91 , 9.32E-3 , 0 , 4.0E+4
820 DATA -271870 , 36.074 , 23.550 , 2.603E-2 , 0 , 1.79E+5
830 DATA -266900 , 37.40 , 24.82 , 2.442E-2 , 0 , 4.0E+4
840 DATA -342340 , 41.975 , 2.876 , 2.379E-2 , 0 , 4.257E+5
850 REM DATA IN 840 GOOD ONLY TO 595C (K2SO4)
890 REM READ IN TEMPERATURES
900 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
910 REM HERE ARE THE STOICHIOMETRIC COEFFICIENTS
920 REM ACID REPLACEMENT REACTIONS
930 DATA 0 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , 0 , -1 , 1 , 0
940 REM NO OH DATA
950 REM NO SILICATE DATA
960 REM OXIDATIVE REPLACEMENT REACTIONS
970 DATA -.5 , -1 , 0 , 0 , 0 , 0,1 , 0 , 0 , -1 , 0 , 1
980 REM NO OH DATA
990 REM NO SILICATE DATA
1000 REM REDOX REACTIONS ON SULFUR
1010 REM NO SULFIDE DATA
1020 REM NO SULFIDE DATA
1030 END
```

```
MG/DAT


780 REM HERE ARE THE-DATA ON MAGNESIUM COMPOUNDS
790 REM MGO , MG(OH) , MGCO3 , MGSO3 , MGSO4 , MGS , MGSIO3
800 DATA -143630 , 6.4025 , 10.175 , 1.7392E-3 , 0 , 1.4788E+5
810 DATA -220820 , 15.077 , 13.034 , 1.579E-3 , 0 , 0
820 DATA -263980 , 15.696 , 18.613 , 1.3794E-2 , 0 , 4.1592E+5
830 DATA -241000 , 22.5 , 17.09 , 1.684E-2 , 0 , 1.48E+5
840 DATA -305500 , 21.9 , 16.53 , 2.18E-2 , 0 , 1.578E+3
850 DATA -83000 , 10.6 , 9.24 , 2.5E-3 , 0 , 7.93E+2
860 DATA -357630 , 16.197 , 24.535 , 5.437E-3 , 0 , 5.867E+5
870 REM
```

```
CA/DAT


160 REM HERE ARE DATA FOR CALCIUM SYSTEM.
170 REM CAO , CA(OH)2 , CACO3 , CASO3 , CASO4 , CAS , CASIO3 ,
180 REM CA3(PO4)2
190 REM NO DATA FOR CAHPO4
200 DATA -151730 , 9.484 , 11.857 , 1.0798E-3 , 0 , 166040
210 DATA -235600 , 19.924 , 25.131 , 2,8822E-3 , 0 , 450780
220 DATA -288110 , 22.194 , 24.965 , 5.237E-3 , 0 , 619700
230 DATA -275880 , 24.201 , 18.770 , 1.618E-2 , 0 , 166000
240 DATA  -340190,25.491,17.220,2.337E-2,0,32560
250 DATA -115300,13.5,10.2,3.8E-3,0,0
260 DATA -377400 , 20.9 , 25.85 , 3.94E-3 , 0 , 565000
270 DATA -986200 , 57.6 , 48.24 , 39.68E-3 , 0 , 500000
280 END
```

BA/DAT

```
790 REM HERE ARE THE DATA ON BARIUM COMPOUNDS
800 REM BAO , BA(OH)2 , BACO3 , BASO3 , BASO4
810 DATA -132070 , 16.795 , 12.733 , 1.0392E-3 , 0 , 198290
820 DATA -226020 , 12.613 , 16.90,21.90E-3, 0,0
830 DATA -287160 , 26.781 , 20.76 , 1.1694E-2 , 0 , 477080
850 DATA -282600 ,28.60 , 19.64 , 1.614E-2 , 0 , 198000
860 DATA -349990 , 31.487 , 33.759 , 3.401E-3 , 0 , 839400
890 REM TEMPERATURES
900 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
910 REM STOICHIOMETRIC COEFFICIENTS
930 REM REPLACEMENT REACTIONS
960 REM OXIDATIVE REPLACEMENTS
970 DATA -.5 , -1 , 1 , 0 , 0 , 0 , 0 , 0 , -1 , 0 , 0 , 1
980 DATA -.5 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , 0 , -1 , 0 , 1
981 DATA 0 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , 0 , -1 , 1 , 0
982 DATA 0 , -1 , 1 , 0 , 0 , 0 , 0 , 0 , -1 , 0 , 1 ,0
990 END
```

MN/DAT

```
790 REM HERE ARE THE DATA ON MANGANESE +2 AND ;3
800REM MNO , MN(CO3) , MNSO3 , MNSO4 , MNS , MNSIO3
810 REM MN2O3 , MN2(CO3)3 , MN2(SO3)3 , MN2(SO4)3
820 REM MANGANOUS
840 DATA -91930 , 14.265 , 11.104 , 1.94E-3 , 0 , 87900
850 DATA -213740 , 20.474 , 21.99 , 9.3E-3 , 0 , 469500
860 DATA -197370 , 24.40 , 18.01 , 1.704E-2 , 0 , 68000
870 DATA -253950 , 26.781 , 29.241 , 8.916E-3 , 0 , 703800
880 DATA -49452  , 18.682 , 11.396 , 1.799E-3 , 0 , 0
890 DATA -302210 , 21.2836 , 26.398 , 38.77E-3 , 0 , 6.156E+5
900 REM MANGANIC
910 DATA -229110 , 26.398 , 24.726 , 8.376E-3 , 0 , 322800
920 DATA -545460 , 44.6 , 41.65 , 5.551E-2 , 0 , 740000
930 DATA -502390 , 53.6 , 45.46 , 5.368E-2 , 0 , 323800
940 DATA -666900 , 61.7 , 52.63 , 6.16E-2 , 0 , 497000
950 REM HERE ARE THE TEMPERATURES
960 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
970 REM
980 REM STOICHIOMETRIC COEFFICIENTS
990 REM MANGANOUS
1000 DATA 0,-1,0,0,0,0,1,  0,-1,1,0,0,0,  0,0,0,0
1010 DATA 0,-1,0,0,1,0,0,  0,0,1,0,0,-1,  0,0,0,0
1020 DATA -.5,-1,0,0,0,0,1,  0,-1,0,1,0,0,  0,0,0,0
```

COMPUTER PROGRAM IV (Cont'd)

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```
MN/DAT CONTINUED


1030 DATA -.5,-1,0,0,1,0,0,  0,0,0,1,0,-1,  0,0,0,0
1040 DATA 0,-2,0,0,0,1,0,  0,0,0,1,-1,0,  0,0,0,0
1050 DATA 0,-1,0,0,0,1.5,0,  2,0,0,0,-2,0  ,0,0,0,0
1060 REM MANGANIC
1070 DATA 0,-1,0,0,0,0,1,  0,0,0,0,0,0,  0,-.3333,.3333,0
1080 DATA -.5,-1,0,0,0,0,1,  0,0,0,0,0,0,  0,-.3333,0,.3333
1090 DATA 0,-2,0,0,0,0,3,  0,0,1,1,0,0,  0,-1,0,0
1100 DATA -.5,-2,0,0,0,0,3,  0,0,0,2,0,0,  0,-1,0,0
1110 DATA 0,-1,0,0,0,0,0, -2,0,0,4,0,0,  0,0,0,-1
1140 END




FE/DAT


790 REM HERE ARE THE DATA ON IRON COMPOUNDS
800 REM FEO , FECO3 , FESO3 , FESO4 , FES , FESIO4
810 REM FE2O3 , FE2(CO3)3 , FE2(SO3)3 , FE2(SO4)3 ,
820 REM FES2
830 DATA -63762 , 14.191 , 11.656 , 7.9993E-3 , 0 , 75970
840 DATA -178550 , 22.194 , 11.625 , 2.678E-2 , 0 , 0
850 DATA -165540 , 25.3 , 18.57 , 17.1E-2 , 0 , 76000
860 DATA -220410 , 25.682 , 20.96 , 1.974E-2 , 0 , 134000
870 DATA -22800 , 15.2 , 12.2 , 2.38E-3 , 0 , 0
875 DATA -343540 , 35.38 , 36.51 , 9.36E-3 , 0 , -670000
880 REM ADDITIONAL DATA AVAILABLE FOR 870 ABOVE L38C
890 REM FERRIC DATA
900 DATA -196420 , 20.88 , 23.48 , 1.859E-2 , 0 , 354800
910 DATA -502660 , 44.8 , 40.4 , 6.872E-2 , 0 , 7.72E+5
920 DATA -457400 , 53.8 , 44.21 , 6.389E-2 , 0 , 355000
930 DATA -615600 , 61.90 , 51.38 , 7.181E-2 , 0 , 529000
940 REM PYRITE
950 DATA -42450 , 12.70 , 18.0 , 1.182E-3 , 0 , 3.12E+5
960 REM READ IN TEMPERATURES
970 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
980 REM STOICHIOMETRIC COEFFICIENTS
990 REM FERROUS, STANDARD FORM 2,2,2
1000 DATA 0,-1,0,0,0,0,1,  0,-1,1,0,0,0,  0,0,0,0,  0
1010 DATA 0,-1,0,0,.5,0,0,  0,0,1,0,0,-.5, 0,0,0,0,  0
1020 DATA -.5,-1,0,0,0,0,1,  0,-1,0,1,0,0,  0,0,0,0,  0
1030 DATA -.5,-1,0,0,.5,0,0,  0,0,0,1,0,-.5,  0,0,0,0,  0
1040 DATA 0,-2,0,0,0,1,0,  0,0,0,1,-1,0,  0,0,0,0,  0
1050 DATA 0,-1,0,0,0,1.5,0,  2,0,0,0,-2,0  ,0,0,0,0,  0
1060 REM FERRIC -- STANDARD FORM 1,1
1070 DATA 0,-1,0,0,0,0,1,  0,0,0,0,0,0,  0,-.3333,.3333,0,  0
```

COMPUTER PROGRAM IV (Cont'd)

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```
FE/DAT CONTINUED


1080 DATA -.5,-1,0,0,0,0,1,  0,0,0,0,0,0,  0,-.3333,0,.3333, 0
1090 DATA 0,-2,0,0,0,0,3,  0,0,1,1,0,0,  0,-1,0,0, 0
1100 DATA -.5,-2,0,0,0,0,3,  0,0,0,2,0,0,  0,-1,0,0,  0
1110 DATA 0,-1,0,0,0,0,0, -2,0,0,4,0,0,  0,0,0,-1,  0
1120 DATA 0,-2,0,0,0,1.5,0  ,0,0,0,1,0,0,  0,0,0,0,  -1
1130 DATA 0,-1,0,0,0,2.5,0, 2,0,0,0,0,0, 0,0,0,0, -2
1140 END
```

```
NI/DAT


790 REM HERE ARE THE DATA ON NICKEL +2 NO +3 DATA
800 REM NIO , NISO3 , NISO4 , NIS ,
810 DATA -57264 , 9.078 , 11.176 , 2.0187E-3 , 0 , -3.8893E-5
820 DATA -155860 , 25.4 , 27.69 , 7.28E-3 , 0 , -58000
830 DATA -218500 , 18.586 , 30.078 , 9.917E-3 , 0 , 0
840 DATA -17480 , 16.10 , 9.245 , 12.795E-3 , 0 , 0
860 REM HERE ARE THE TEMPERATIRES
870 DATA 298.15,400,500,600,700,800,900,1000
880 REM HERE ARE THE STOICHIOMETRIC MULTIPLIERS (0,0,2)
890 DATA 0 , -2, 0, 0 , 0 , 1 , 0 , 0 , 0 , 1 , -1
900 DATA 0 , -1 , 0 , 0 , 0 , 1.5 , 0 , 2 , 0 , 0 , -2
910 END
```

```
CO/DAT


790 REM HERE ARE THE DATA FOR COBALT
800 REM COO , COCO3 , COSO3 , COSO4 , COS --NO CO+3
810 DATA -57073 , 12.655 , 11.534 , 2.042E-3 , 0 , -39896
820 DATA -173300,21.99 ,17.17 , 1.875E-2 , 0 , 99000
830 DATA -161750 , 25.5 , 18.44 , 1.714E-2 , 0 , -40000
840 DATA -207340 , 27.067 , 30.095 , 9.915E-3 , 0 , 691.5
850 DATA -22760 , 16.18 , 10.5952 , 2.51E-3 , 0 , 0
870 REM TEMPERATURES
880 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
930 REM REPLACEMENT REACTIONS
940 DATA 0 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , -1 , 1 , 0 , 0
```

```
CU/DAT CONTINUED


950 REM OXIDATIVE REPLACEMENTS
960 DATA -.5 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , -1 , 0 , 1 , 0
970 REM REDOX ON SULFUR
980 DATA 0 , -2 , 0 , 0 , 0 , 1 , 0 , 0 , 0 , 0 , 1 , -1
990 DATA 0 , -1 , 0 , 0 , 0 , 1.5 , 0 , 2 , 0 , 0 , 0 , -2
```

```
CU/DAT


790 REM HERE ARE THE DATA ON COPPER COMPOUNDS
800 REM CUO , CUCO3 , CUSO3 , CUSO4 , CUS
805 REM CU20 , CU2CO3 , CU2SO3 , CU2SO4 , CU2S
810DATA  -39490 , 10.187 , 9.267 , 4.797E-3 , 0 , 0
820 DATA -142100 , 21.223 , 14.91 , 2.151E-2 , 0 , 139000
830 DATA -127280 , 25.70 , 16.18 , 1.99E-2 , 0 , 0
840 DATA -184220 , 27.067 , 18.805 , 1.718E-2 , 0 , 2792
850 DATA -11600 , 15.9 , 10.6 , 2.64E-3 , 0 , 0
855 REM CUPROUS
860 DATA -40780 , 22.521 , 14.893 , 5.698E-3 , 0 , 0
870 DATA-142290 , 37.63 , 20.53 , 2.241E-2 , 0 , 1.39E+5
880 DATA -127000 , 40.60 , 21.8 , 2.05E-2 , 0 , 0
890 DATA -179510 , 38.465 , 24.19 , 23.4E-2 , 0 , 58000
900 DATA -19.931 , 28.883 , 19.492 , 0 , 0 , 0
910 REM CU2S DATA (900) APPLY ONLY TO THE 25 TO 103 C RANGE
911 REM TEMPERATURES
912 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
920REM STOICHIOMETRIC COEFFICIENTS
930 REM STD SEQUENCE FOR CUPRIC 1,1,2
940 DATA 0, -1, 0, 0, 0, 0, 1,   0, -1, 1, 0, 0,   0, 0, 0, 0, 0
950 DATA -.5, -1, 0, 0, 0, 0, 1,   0, -1, 0 ,1, 0,   0, 0, 0, 0, 0
960 DATA 0, -2, 0, 0, 0, 1, 0,   0, 0, 0, 1, -1,   0, 0, 0, 0, 0
970 DATA 0, -1, 0, 0, 0, 1.5, 0,   1, 0, 0, 0, -2,   0, 0, 0, 0, 0
980 REM STD SEQUENCE FOR CUPROUS
990 DATA 0, -1, 0, 0, 0, 0, 1,   0, 0, 0, 0, 0,   0, -1, 1, 0, 0
1000 DATA -.5, -1, 0, 0, 0, 0, 1,  0, 0, 0, 0, 0,  0, -1, 0, 1, 0
1010DATA 0, -2, 0, 0, 0, 1, 0,  0, 0, 0, 0, 0,  0, 0, 0, 1, -1
1020 DATA 0, -1, 0, 0, 0, 1.5, 0,  0, 0, 0, 0, 0, 1, 0, 0, 0, -2
1030 REM REDOX INVOLVING COPPER
1040 DATA 0, -1, 0, 0, 0, 0, 2, 0, -2, 0, 0, 0, 0, 0, 0, 1, 0
1050 END
```

COMPUTER PROGRAM IV (Cont'd)

```
SN/DAT


790 REM HERE ARE THE DATA ON TIN +2 AND +4
800 REM STANNOUS SNO , SNS
810 DATA -68325 , 13.555 , 9.5464 , 3.4975E-3 , 0 , 0
820 DATA -24320 , 18.395 , 8.4795 , 7.428E-3 , 0 , 95260
830 REM STANNIC SNO2 , SNS2 , SN(CO3)2 , SM(SO3)2,SN(SO4)2
840 DATA -138750 , 12.504 , 17.652 , 2.3986E-3 , 0 , 515790
850 DATA -39896 , 20.880 , 15.510 , 4.197E-3 , 0 , 96.3
860 DATA -330040 , 33.9 , 28.94 , 3.58E-2 , 0 , 794000
870 DATA -295360 , 42.9 , 31.48 , 3.26E-2 , 0 , 5.16E+5
880 DATA -393400 , 37.1 , 36.26 , 3.788E-2 , 0 , 632000
885 REM HERE ARE THE TEMPERATURES
890 DATA 298.15 , 400 , 500 , 600 , 700 , 800 , 900 , 1000
900 REM STOICHIOMETRIC COEFFICIENTS
910 REM STANNOUS
920 DATA 0 , -1 , 0 , 0 , 0 , 1.5 , 0 , 2 ,-2 , 0 , 0 , 0 , 0 , 0
930 REM STANNIC
940 DATA 0 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , 0 , 0 , 0 , -.5 , .5 , 0
950 DATA -.5 , -1 , 0 , 0 , 0 , 0 , 1 , 0,0, 0 , 0 , -.5 , 0,.5
960 DATA 0 , -2 , 0 , 0 , 0 , 1 , 0 , 0 , 0 , 0 , -.5 , 0 , 0 , .5
970 DATA 0 , -1 , 0 , 0 , 0 , 1.5 , 0 , 0 , 0 , 1 , -1 , 0 , 0 , 0
980 END
```

```
ZN/DAT


790 REM HERE ARE THE DATA ON ZINC COMPOUNDS
800 REM ZNO , ZNCO3 , ZNSO3 , ZNSO4 , ZNS
810 DATA -83376 , 10.392 , 11.704 , 1.2184E-3 , 0 , 217880
820 DATA -193680 , 19.685 , 9.2932 , 3.2968E-2 , 0 , 0
830 DATA -178480 , 25.8 , 18.610 , 2.728E-2 , 0 , 2.18E+5
840 DATA -233690 , 29.767 , 17.062 , 2.6791E-2 , 0 , 0
850 DATA -48500 , 13.8 , 12.16 , 1.236E-3 , 0  , 1.361E+5
860 REM HERE ARE THE TEMPERATURES
870 DATA 298.15 , 400 , 500 , 600  ,700 , 800 , 900 , 1000
880 REM HERE ARE THE STOICHIOMETRIC COEFFICIENTS
900 DATA -.5 , -1 , 0 , 0 , 0 , 0 , 1 , 0 , -1 , 0 , 1 , 0
910 DATA 0 , -2 , 0 , 0 , 0 , 1 , 0 , 0 , 0 , 0 , 1 , -1
930 END
```